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CONSIDERATIONS REGARDING THE OCCURRENCE OF RADON, THORON, AND THEIR DECAY PRODUCTS IN NON-URANIUM MINES

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ABSTRACT

The occurrence of radon, thoron and their short-lived decay products in non-uranium mines is examined. The basic physico-chemical mechanisms responsible for the exhalation of radon and thoron from soils and rock formations, and their pathways to living and working environments are reviewed. Furthermore, expressions are given that allow the calculation of radiation levels in non-uranium mines from basic information and fundamental principles. Finally, information on radiation levels (radon and its decay products) in non-uranium mines in Canada and other countries is given followed by a brief description of radiation control methods in underground mines.

Key words: Radon and radon daughters; Thoron and thoron daughters; Underground mines.

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RÉSUMÉ

Nous nous penchons dans le présent document sur la présence de radon et de thoron ainsi que de leurs produits de désintégration éphémères dans les mines autres que les mines d'uranium. Sont étudiés les mécanismes physico-chimiques fondamentaux à l'origine de l'émanation du radon et du thoron par les sols et les formations rocheuses, ainsi que les voies par lesquelles ils parviennent dans les milieux de vie et de travail. Sont aussi données des expressions permettant de calculer les niveaux de rayonnement présents dans les mines autres que les mines d'uranium à partir de renseignements et de principes fondamentaux. Enfin, nous donnons des renseignements sur les niveaux de rayonnement (du radon et de ses produits de désintégration) présents dans les mines autres que les mines d'uranium du Canada et d'autres pays, ainsi qu'une courte description des méthodes de radioprotection utilisées dans les mines souterraines.

Mots clés : radon et descendants du radon; thoron et descendants du thoron; mines souterraines

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INTRODUCTION

Radioactive elements are a common occurrence on earth and in our society. A radioactive element is a chemical element, i.e., atomic species, whose nucleus is in an excited energy state, and hence physically unstable. Because of this, the atom decays to a more probable, lower energy state by emitting the excess of energy in the form of electromagnetic radiation (e.g., γ-rays) or particulate matter (e.g., α- and β-particles). Radioactive elements (or radioisotopes or radionuclides, as they are most commonly known), constitute a source of energy which poses a potential health hazard by interacting with living matter, i.e., tissue. The most common interaction between radiation and tissue is by ionization.

Radionuclides can be of man-made origin or naturally occurring. Usually, radionuclides are part of both simple and complex organic and inorganic molecules forming a variety of chemical compounds.

Many radionuclides of man-made origin are widely used for commercial, industrial, engineering, medical and research purposes. The majority of man-made radionuclides are produced in nuclear reactors by neutron irradiation of certain materials or in particle accelerators by a variety of high-energy particle collisions.

Radionuclides of man-made origin (e.g., ^{137}Cs, ^{131}I, and many others) are also found in the biosphere and as contamination of the surface of the earth in livestock, fish, foods, water supplies. milk, grass, trees, crops, and the like, as a result of nuclear detonations (atomic and hydrogen bombs), and of nuclear reactor accidents, such as Three-Mile Island (U.S.A.) and Chernobyl (U.S.S.R.) (1-3).

Naturally-occurring radioisotopes (e.g., ^{238}U, ^{235}U, ^{232}Th, and their decay products, and ^{40}K) are found in the earth in several geological formations and locations such as the continental and oceanic crusts and the mantle beneath the oceans (4). These radioisotopes have been in the earth

ever since its formation a few billion years ago. Naturally occurring radionuclides can also be of extraterrestrial origin, e.g., meteoritic origin (5). The bombardment of meteorites by cosmic radiation over long periods of time in their journey through space results in the production of several radioisotopes rare in nature such as ^{3}H, ^{53}Mn, ^{10}Be, and ^{26}Al. Other radionuclides include ^{129}Xe, and uranium and thorium radioisotopes. These radioisotopes of meteoritic origin are found on the earth's surface when meteorites enter the earth's atmosphere and 'disintegrate' by friction or collide on the earth's surface.

In addition to the above, the earth is continuously bombarded by cosmic radiation of solar, galactic and extragalactic origin (6,7). In this case, however, the chief components of cosmic radiation are not radionuclides, but (mainly) highly energetic charged particles such as protons, α-particles, electrons, and muons, and highly penetrating electromagnetic radiation such as γ-rays and X-rays. A great deal of the cosmic radiation penetrating the earth's atmosphere is associated with the sun's activity, i.e., solar flares.

Biological systems (i.e., human, animals and the vegetal world) are, therefore, exposed to a variety of radiation sources both man-made and naturally occurring. It has recently been recognized that radon (^{222}Rn) and its short-lived decay products (daughters or progeny as they are commonly known) are the most important single contributor to human exposure to radiation, man-made or naturally occurring (3).

Radon is formed by the radioactive decay of ^{238}U (uranium) through a series of intermediate radioactive decay steps. This radioisotope is the parent of a naturally occurring radioactive chain known as the ^{238}U-decay chain. Because of the importance of ^{222}Rn and its short-lived decay products from the health physics and occupational standpoints, main emphasis will be placed in this paper on the occurrence of these radionuclides at the workplace and at home.

It is worth bringing to the attention of the reader that some interest has recently been focused on another radioactive gas, namely, thoron (^{220}Rn) and its short-lived decay products as significant potential contributors to radiation exposure in humans. Thoron arises from the radioactive decay of another naturally-occurring radioactive decay chain, whose parent is thorium (^{232}Th).

ORIGIN OF THE NATURAL RADIOACTIVE DECAY CHAINS AND OTHER RADIONUCLIDES ON THE EARTH CRUST

There are a number of radionuclides found on earth of natural origin (8). Table 1 shows a list of natural radionuclides of half-life ($T_{1/2}$) greater than 10^8 y. All long-lived naturally-occurring radionuclides found on earth after its formation have been formed elsewhere in stellar nucleosynthesis or supernova explosions.

Very heavy radionuclides, i.e., high atomic and mass numbers radioisotopes such as ^{235}U, ^{238}U, ^{226}Ra and ^{232}Th have been formed in the last stages of stellar evolution, such as supernova explosions, as follows: (a) At the very high temperatures generated in the collapse and explosion, some of the nuclei in massive stars are broken up, and many neutrons and protons are formed. The neutrons and protons are captured by other nuclei, building up heavier elements such as silver (Ag), gold (Au), uranium (U), and thorium (Th). In this way, elements of the periodic table extending beyond iron (Fe) are manufactured in the final moments of the massive star's life, i.e., they are mostly created in the brief moments of the supernova explosion. Because the time available for making these heavy elements is so brief, they never become as abundant as the lighter elements, i.e., up to and including Fe. The radioactive substances ^{226}Ra, ^{235}U, ^{238}U, ^{232}Th and ^{40}K, among others, have very large ionic radii. Because of this, these elements are concentrated in the earth's crust by differentiation, that is, removal from the mantle to the crust.

The debris resulting from supernova explosions are scattered to vast distances in all directions in outer space. Some of this debris contains the long-lived radionuclides indicated above. Since this debris (supernova remnants) form part of interstellar matter from which stars and planets are formed, it is not difficult to visualize, and predict, the occurrence of heavy, long-lived radionuclides on the surface of the earth. (It should be noted that the most widely accepted theory on the birth of the solar system is a modified version of the Laplace theory in which the sun forms at the centre of a rotating gas disc, with the planets condensing from rings of matter further out.)

In summary, a number of long-lived radionuclides are found on earth. Three of these radionuclides are the parents of the three natural radioactive decay chains, namely, ^{235}U, ^{238}U and ^{232}Th. Some important radionuclides are shown in Table 1, other radionuclides (not shown in Table 1) are found in meteoritic remnants.

Because of its importance in the context of this discussion, the detailed radioactive decay schemes of the three naturally-occurring radioactive chains are shown in Figures 1-3. In all other cases, the important radioactive elements from the health physics and occupational standpoints are ^{222}Rn, ^{220}Rn, and their respective short-lived decay products. These radionuclides are shown in detail in Figures 4 and 5.

DISTRIBUTION OF THE ELEMENTS AND THEIR PROPERTIES

The eleven most abundant elements in the earth's crust are given in Table 2 (10). These elements make up about 99.3% in weight of all known elements in the earth's crust. Many other very important elements found on earth such as vanadium (V), chromium (Cr), titanium (Ti), manganese (Mn), cobalt (Co), nickel (Ni), copper (Cu), zinc (Zn), niobium (Nb), molybdenum (Mo), silver (Ag), cadmium (Cd), tin (Sn), barium (Ba), tungsten (W), platinum

(Pt), gold (Au), mercury (Hg), lead (Pb), thorium (Th), and uranium (U), are found in either low abundance or as trace elements.

It is quite surprising that many elements such as Sn, Cu, Zn, Pb, Hg, Ag, Pt, Sb, As, Au and U, that are so essential to our needs and culture, are among some of the rarest elements in the earth's crust.

Uranium, although considered a trace element is widely distributed in all kinds of rocks and in the sea. There are on average about 3 parts per million by weight in the earth's crust (3 g/tonne), and about 3 parts per billion by weight in the sea, which corresponds to a total mass in the ocean of ~4 x 10^9 Kg. In spite of this seemingly low abundance, uranium is about 10^3 times as abundant as gold (Au), and is as common as, or more abundant than, a number of other minerals such as silver (Ag), cadmium (Cd), mercury (Hg), platinum (Pt), molybdenum (Mo), tungsten (W), arsenic (As), and antimony (Sb).

Thorium is also widely distributed in the earth's crust and even more abundant than uranium. The gram ratio Th/U varies widely, but can be considered according to experimentation as $\geq$ 3.0 - 4.0.

Naturally occurring uranium contains the following radioisotopes and percentages:

^{238}U : 99.28%

^{235}U : 0.71%

^{234}U : 0.0058%

The radioisotope ^{235}U is a naturally occurring fissionable material. Uranium-235 can be separated from ^{238}U by gaseous diffusion. Uranium is a heavy, silvery white metal which is pyrophoric when finely divided.

Thorium-232 is a dark-gray metal. Thorium is fissionable and is a potential source of nuclear power. Thorium can be used as a nuclear fuel for nuclear reactors of the breeder type. However, this metal and its different chemical forms have not received the attention of uranium, due to the demand

for plutonium (Pu). It has been suggested that there is probably more energy available for use from Th in the minerals of the earth than from both U and fossil fuels.

The elements (metals) U and Th are rarely found in element form in nature, but mostly combined with other chemical elements to form simple and complex chemical compounds. Some useful properties of U and Th as well as other relevant elements are shown in Table 3.

Uranium is commonly found in combined chemical form in such minerals as pitchblende, uraninite, carnotite, autunite, uranophane, davidite, brannerite, and tobernite. The most common chemical forms of thorium are thorite ($ThSiO_4$) and thorianite ($ThO_2 + UO_2$).

Uranium is found in large concentrations suitable for economic mining in several parts of the world such as Canada, U.S.A., South Africa and Australia.

Uranium compounds are found in a variety of common rocks and sediments of the type, and in concentration as indicated below (11):

Peridotite, pyroxenite, dunite, and other ultramafics .. 0.03 ppm

Gabbro, basalt, and other mafics ..... 0.05 ppm

Granite, granodiorite, and other felsics ..... 3.50 ppm

Andesite, diorite, and other intermediates ..... 1.80 ppm

Shale, clay, and other sedimentaries ..... 3.20 ppm

Although uranium occurs widely on all five continents, the highly concentrated deposits necessary for economic mining are uncommon. Mineable concentrations range from 4×10^2 to $>3 \times 10^3$ times the average crustal abundance of ~3 ppm. The favourable rocks and geologic environments are limited to a few major types (11), namely:

1. Uraniferous vein deposits;
2. Uranium deposits in quartz-pebble conglomerates;
3. Uraniferous igneous rock;

4. Uraniferous marine black shales;

5. Uraniferous phosphatic rocks;

6. Peneconcordant uraniferous deposits; and

7. Other epigenetic uraniferous deposits.

Over 95% of the reported uranium reserves occur in six metallogenic regions (12):

Colorado Plateau and Wyoming (sandstones) (U.S.A.)

Witwatersrand Basin (South Africa)

Northern Saskatchewan and Ontario (Canada)

Northern Territory (Australia)

Niger, Gabon (Central Africa)

European Hercynian belts.

Other important deposits include Rössing (South West Africa) and in South Australia.

Uranium minerals are often found with other mineral compounds in varying amounts. Minerals which have been found to be 'associated' with uranium compounds include Th, Zr, Cu, As, Mo, Pb, Ca, Ag, Zn, Ti, Co, Ni, V, and Fe. The ratio Th/U in some regions, e.g., Elliot Lake/Blind River, ranges between 1:4 to 4:1.

Radon is commonly regarded as a totally chemically inert gas. However, radon has been shown (13) to be chemically active to a surprising degree. A considerable amount of work has been carried out on the chemistry of radon by Stein (13, and references therein), and others (14, 15).

Radon is a metalloid (13), i.e., an element that is situated on the diagonal of the periodic table between the metals and non-metals. Because of its position in the periodic table, radon exhibits some of the chemical characteristics of both, metal and non-metal.

Radon reacts with fluorine (F), halogen fluorites, dioxygenyl salts, fluoronitrogen salts, and halogen fluorite-metal fluoride complexes to form

ionic compounds. Solutions of non-volatile, cationic radon have also been produced in non-aqueous solvents. Radon chemical compounds, however, hydrolyze easily and must be protected against moisture. Radon can be quantitatively collected on ion-exchange columns packed with Nafion resins or complex salts. Application of several chemical compounds such as $IF_6^+SbF_6^-$ and $O_2^+SbF_6^-$ for trapping radon in uranium mines and other radon laden working environments have been suggested by Stein et al. (13). Some useful properties of radon are given in Table 3.

Apart from the radioactivity properties of thoron, such as half-life, virtually nothing is found in the literature regarding the chemistry and chemical properties of thoron. This is most probably due to the short half-life of this radioactive gas (~55 s) which precludes its detailed study and severely limits its handling to short periods of time.

Data pertaining to radium and thorium concentrations in coal and coal residues for different geographical locations and coal or residue materials are given in Table 4. Average radon gas concentrations in natural gas at the well and in the distribution line for several countries are given in Table 5.

EMANATION AND MIGRATION OF RADON AND THORON IN THE GROUND

As previously discussed, radon (^{222}Rn) and thoron (^{220}Rn) are produced by the radioactive decay of ^{238}U and ^{232}Th, respectively. However, there is no simple correlation between the parents of the two naturally occurring radioactive chains and their respective radioactive gases. This is so because the immediate precursors of ^{222}Rn and ^{220}Rn are ^{226}Ra and ^{224}Ra, respectively, which in turn are derived from other (parent) radionuclides. Because the chemical characteristics of the long-lived radionuclides ^{238}U, ^{234}U, ^{230}Th and ^{226}Ra are so different, they are frequently separated by a variety of geochemical processes (16). For example, ^{238}U is generally immobile in reducing environments and mobile in oxidizing environments, whereas the

movement of ^{234}U is much less restricted by the mass-action law. (If radioactive equilibrium in the radioactive chain prevails, the concentration of ^{234}U would be a factor of about 2×10^4 lower than that of ^{238}U. In fact, this factor could be significantly greater because radioactive equilibrium does not usually prevail for the reasons indicated above, namely, the different geochemical processes affecting the members of the chain.) Thorium-230 is the least mobile of the long-lived radionuclides of the chain probably because of its adsorption on silicates. Radium-226 is fairly mobile in reducing environments. However, in oxidizing environments ^{226}Ra is coprecipitated with alkaline-earth compounds, particularly $CaCO_3$, or can be scavenged by oxidizing Fe or Mn. Uranium, and radium and its progeny (i.e., ^{222}Rn and its short-lived decay products) are effectively separated in natural waters. Hence, measurement of ^{226}Ra in water gives little information about the uranium distribution in the area, and vice versa.

Similar arguments as those discussed above could be applied to the long-lived radionuclides ^{232}Th, ^{228}Ra, ^{228}Th and ^{224}Ra which are members of the naturally occurring thorium radioactive chain.

It should be noted that although qualitatively speaking a correlation can be found in many cases between the occurrence of ^{226}Ra and the measurement of ^{222}Rn in the same general area, the correlation is not a straightforward one from the quantitative point of view. This is so because factors such as geological formation, ground characteristics, hydrology and meteorological conditions play such a major role. Any correlation between ^{224}Ra and ^{220}Rn is even more obscure than between ^{226}Ra and ^{222}Rn because of the short radioactive half-life of ^{220}Rn (54.5 s), and hence, short diffusion length.

The occurrence of ^{222}Rn (and also to a significant extent ^{220}Rn) in living and working environments is of great concern from the individual health risk assessment and personnel occupational standpoints.

It is clear that the migration of ^{222}Rn and ^{220}Rn in soil (and water)

and their entry into living and working locations require the following requisites to be satisfied:

1. there must be ^{226}Ra and ^{224}Ra in the ground;
2. the ^{222}Rn and ^{220}Rn formed by the decay of ^{226}Ra and ^{224}Ra, respectively, should also be able to migrate to the structure, e.g., house, plant, factory, underground mine, mill, or any other mining or processing installation;
3. Pathways for ^{222}Rn and ^{220}Rn to migrate to the living or working location should be available;
4. Favourable meteorological conditions should prevail, e.g., higher barometric pressure in the ground than in the location of interest.

Items 2 to 4 will be discussed next. Item 1 will be assumed as a prerequisite, and hence will not be dealt with here. The reader should be aware, however, that the occurrence of ^{222}Rn at a given location could conceivably, and not uncommonly, arise from ^{226}Ra sources situated at great distances from the location in question. In this case, the hydrological conditions of the area play a crucial role as ^{222}Rn, which is readily dissolved in water, could be transported by the latter to the location of interest provided a suitable pathway is available.

MECHANISMS OF ^{222}Rn RELEASE FROM SOLIDS

Uranium and thorium bearing rocks are broken down by thermal expansion and contraction, water erosion and chemical action, earth movement, wind, and other forces and mechanisms. These natural forces result in products ranging from large blocks or aggregates to fine particles and solutions. The space between these solid fragments, i.e., pores, are filled with fluids such as air and/or water. The porosity of the medium and the fluids in the pores are important factors which effectively determine the escape of ^{222}Rn from solids, and its migration in the ground.

There are several release mechanisms of ^{222}Rn from solids. It is

important in this context to recall that ^{222}Rn is ejected from the decay of ^{226}Rn by α-recoil mechanisms, i.e., $^{226}Ra \rightarrow {}^{222}Rn + \alpha$. Mass and momentum conservation laws demand ^{222}Rn and the α-particle to be emitted in exactly opposite directions. This process is isotropic. The energy of the recoil particle varies with the decaying radionuclide but is about 0.1 MeV. The recoiling ^{222}Rn atom has a range (R) related to its energy and the medium through which it travels, i.e., rock, air, water (17-19).

Depending on the location of the ^{226}Ra atom in the solid fragment, and the physical dimensions of the fragment, the following mechanisms, shown in Figures 6 and 7, are operative (assume spherical fragments of diameter D_p, for simplicity) (20-22):

1. If $D_p >> R$ and the ^{226}Ra atom is situated at a distance, d, from the surface of the solid fragment such that $d>R$, then the recoiling ^{222}Rn atom passes through crystalline material, and bores a small tunnel that quickly fills with the solidified material that was temporarily 'melted' and pushed out of its path (20). Radon atoms thus sealed into the crystal structure of the solid fragment usually decay before they have time to diffuse to the crystal surface. Sometimes, however, if the recoiling ^{222}Rn atom is not too far from the fragment surface it can emerge to the interstitial space by diffusion mechanisms, i.e., Fick's law.
2. If $D_p >> R$, but the ^{226}Ra atom is situated at a distance from the surface of the solid fragment such that $d<R$, then there is a chance for the recoiling ^{222}Rn to emerge into the pore space provided the α-particle is emitted in the radial direction, i.e., ^{222}Rn atoms ejected outwards along the radius of the solid fragment.
3. If $D_p >> R$, but the ^{226}Ra is situated on the surface of the solid fragment there is a larger probability for the recoiling ^{222}Rn atom to enter the pore space than to be ejected into the solid fragment.
4. If $D_p < R$, the recoiling ^{222}Rn will escape the solid fragment and will

emerge in the pore space.

The range of the recoiling ^{222}Rn atom depends on the composition and density of the matter of the medium it encounters. The range in water and air is about 0.05 μm and 50 μm, respectively, whereas in a solid with the elemental composition of coal the range is about 0.04 μm (23). Alternatively, the ranges in, say, water, air, silica, bituminous coal and lignite are 5.2, 6.1, 6.3, 5.4 and 5.5 $\mu g/cm^2$, respectively (22). Hence, if any of these materials are mixed intimately on a scale compared to the recoil distance (R), the probability of the ^{222}Rn atom stopping in a particular material is approximately proportional to the mass fraction of that material in the composite (22).

In addition to the above mechanisms of ^{222}Rn release from solids there is another, less direct mechanism which occurs when the pore space is filled with air and the distance between adjacent solid fragments is less than the range of the recoiling ^{222}Rn atom. This case could apply to items 2 to 4, namely, the recoiling ^{222}Rn atom leaves the surface of a solid fragment, traverses the air filled pore space, and is embedded in the opposite solid fragment. The embedded ^{222}Rn atoms produce radiation-damaged regions as they enter the new solid fragment. If water later reaches the damaged region, it preferentially dissolves the damage and may release whatever embedded ^{222}Rn has not already decayed (21).

Radon atoms that stop in liquids in the pore space, e.g., water, are free to diffuse, and if the liquid provides a path to a free surface that is not too long, ^{222}Rn atoms can escape from the solid fragment or aggregate before they decay. For example, the diffusion length (defined below) of ^{222}Rn in water (in the liquid state) is 2.3 cm. A simple calculation shows that a recoiling ^{222}Rn atom has an 80% probability of diffusing through 0.5 cm of liquid water before radioactive decay occurs, but only about 11% proability through 5 cm (24). Radon atoms that stop in air in interstitial spaces

(pores) are also free to diffuse. However, as examined below, there are other mechanisms of ^{222}Rn escape from interstitial spaces to nearby and far removed locations.

MECHANISMS OF ^{222}Rn AND ^{220}Rn ESCAPE FROM SOILS AND FRAGMENTED SOLIDS

Once ^{222}Rn is in interstitial spaces it can find its way to other locations by three basic mechanisms, namely:

1. A radioactive gas (^{222}Rn and ^{220}Rn) concentration gradient-induced or diffusion mechanism governed by Fick's Law.
2. A pressure-induced or convective transport mechanism governed by Darcy's Law.
3. Transport of ^{222}Rn and ^{220}Rn by liquid water movement through pore spaces, capillaries, and the soil and rock formations.

The above mechanisms (shown in Figure 8) are greatly influenced, and usually enhanced, by fissures, cracks, and faults in the soil and rock formations.

In its simplest form, Fick's Law can be written as:

$$J_D = -D\nabla C \qquad \text{Eq 1}$$

where, J_D is the current density (flux) of radioactive atoms by molecular diffusion. The symbol D represents the bulk diffusion coefficient of the radioactive gas (^{222}Rn, ^{220}Rn) in the medium (soil, etc.), whereas ∇C represents the concentration gradient.

Darcy's Law of convective transport can be expressed as:

$$J_t = C\upsilon \qquad \text{Eq 2}$$

where,

$$\upsilon = -(k/\delta)\nabla P \qquad \text{Eq 3}$$

where, in Equations 2 and 3:

J_t is the current density (flux) of radioactive atoms by convective transport;

C is the radioactive gas concentration;

υ is the fluid volume current density (given by Darcy's Law, Equation 3),

of interstitial fluid flowing per unit time per unit geometrical area for pressure-induced flow;

k is the soil permeability;

δ is the dynamic viscosity of interstitial fluid;

VP is the pressure gradient.

The total ^{222}Rn or ^{220}Rn flux, J_T, is given from Equations 1 and 2, as:

$$J_T = J_D + J_t \qquad \text{Eq 4}$$

It should be noted that in the context of this discussion, item 2 is meant to apply to convective transport of radioactive gas by air in interstitial spaces, whereas item 3 is meant to relate to bulk transport of the radioactive gas by ground water and surface water on a macroscopic, i.e., large scale.

As discussed in more detail in Appendix A, Fick's and Darcy's Laws can be applied to the formulation of explicit expressions for the flow of radioactive gas through any medium and media interface such as mine walls, tailings piles, open pit mines, and ground/home interfaces.

The reader should be aware that the contribution of a source of ^{222}Rn or ^{220}Rn to a given location is limited by the radioactive half-life of these gases. Hence, the porosity and permeability of the medium as well as the diffusivity (i.e., diffusion coefficient) of the radioactive gas in the medium and the velocity of air and/or water (in pressure-induced transport mechanisms) in the medium are very important variables. A useful variable for concentration gradient-induced mechanisms is the so-called diffusion length, L, defined as:

$$L = (D/\eta\lambda)^{1/2} = (D\tau/\eta)^{1/2} \qquad \text{Eq 5}$$

where, η is the porosity of the medium and λ is the radioactive decay constant of the radioactive gas, i.e., $\lambda(^{222}Rn) = 2.1 \times 10^{-6}\ s^{-1}$ and $\lambda(^{220}Rn) = 1.27 \times 10^{-2}\ s^{-1}$. The symbol τ is $\tau = 1/\lambda$. Diffusivity, diffusion length, porosity and permeability data for several media for ^{222}Rn and ^{220}Rn are given in Table

6. (Notice that L_{max} ~5 m.)

For pressure-induced mechanisms (i.e., convective transport), the transport velocity v in conjunction with $\lambda(=1/T_{1/2})$ determines the contribution of radioactive gas sources to a given location. The emanation of ^{222}Rn and ^{220}Rn from (and diffusion through) different media depends on the medium moisture content and temperature, and grain size, among other factors (21, 22, 25-28).

RADON AND THORON GASES IN UNDERGROUND MINES

The release, escape and migration mechanisms examined above (see also Figures 8 to 10, and Appendix A) are responsible for the occurrence of radon and thoron, and their respective progenies, in underground uranium and non-uranium mines, tailings piles, open pit mines, and other areas of interest such as homes. Although the main emphasis is placed on underground non-uranium mines, the same or similar arguments as those used below are applicable to other cases of practical interest such as those indicated above.

For non-uranium mines, there are three cases which are of interest in the context of this paper:

1. Uranium, thorium and radium content in mine walls is vanishingly low and hence negligible for all practical purposes;
2. Uranium, thorium and radium content in mine walls is appreciable, and 'associated' with other metal compounds to be extracted; and
3. Either one of items 1 or 2 but appreciable amounts of ground and/or surface water also reaches mine openings through cracks, fissures, bore holes, rock faults, or any other means or pathways.

If item 1 is applicable, and no surface and/or ground water reaches mine openings, one may safely assume ^{222}Rn and/or ^{220}Rn concentration levels in the mine to be negligible. However, if water finds its way into the mine, potentially high concentration levels of ^{222}Rn are possible. This is based on

the assumption that water goes through a region(s) where uranium, thorium and/or radium content is appreciable. Hence, ^{222}Rn in interstitial pore spaces is dissolved in water and subsequently carried by it into mine openings. Because of the short half-life of ^{220}Rn, no appreciable amount of this radioactive gas is expected to reach mine openings by the above mechanism. If water does not encounter regions of appreciable uranium, thorium and/or radium concentration in its path, ^{222}Rn concentration levels in the mine should be expected to be very low or non-existent. The case of an underground non-uranium mine with negligible U, Th and Ra content in its walls, and no water entry, will be referred to as case A. The situation for the same type of mine but with water entry will be referred to as case B.

The case portrayed in item 2, with no water entry, will be referred to as case C, whereas the same case but with water entry into mine openings will be referred to as case D. Case A is of no interest here. Cases B, C, and D will be briefly discussed next.

Radioactivity conditions in underground uranium mines (e.g., dynamic behaviour of radon and its progeny) has been considered by several authors who have proposed a number of radiation mine models (29-33). An extension of these models to thoron and its progeny, and to time-dependent conditions for both radon and its progeny and thoron and its progeny has also been investigated (34-37). The same basic principles and formulation can be applied directly to 'non-uranium' mines as indicated below.

CASE B - MINE WITH NO U, Th, AND Ra CONTENT IN WALLS AND WATER ENTRY

In this case, only discrete sources of ^{222}Rn need to be considered (i.e., water entry locations) as opposed to a uniformly extended ^{222}Rn source as it is usually assumed in underground uranium mine models (see Figure 9). The solutions to the set of differential equations describing the growth of ^{222}Rn and its progeny reduces, in this case, to considering only the constants appearing in the pertinent solutions for the case of an underground uranium

mine. These constants are reproduced in Appendix B for the benefit of the reader.

CASE C - MINE WITH APPRECIABLE U, Th AND Ra CONTENT IN WALLS AND NO WATER ENTRY

This case (see Figure 10) is not distinguishable from the case of a 'dry' uranium mine. The solutions to the equations describing the growth of ^{222}Rn, ^{220}Rn and their respective progenies are identical and will not be duplicated here. The reader is advised to consult the pertinent literature on the subject (29-37).

CASE D - MINE WITH APPRECIABLE U, Th, AND Ra CONTENT IN WALLS AND WATER ENTRY

This case can be dealt with as a combination of cases B and C. Since the equations describing radioactivity growth for these two cases are linearly independent, each case can be solved independently and the results added together.

It is worthwhile mentioning, for the sake of completeness, that the ^{222}Rn discrete source case (e.g., case B and, in part, case D) could be approached by assuming an 'equivalent', average, radioactive gas flux, i.e., extended source case, and then using the conventional underground uranium mine model equations.

INHALATION OF RADON, THORON, AND THEIR SHORT-LIVED DECAY PRODUCTS

When radon is inhaled, part of it will dissolve in body fluids and get into the blood stream where it, and the short-lived decay products formed during its stay inside the body, will lead to the irradiation of tissue and several organs. However, because of the reported chemical inertness of ^{222}Rn, and its short retention time in the respiratory system during breathing as compared to its radioactive half-life (3.82 d), the potential health hazards associated with radon per se are assumed to be rather low.

The case for thoron is somewhat different to that of radon because of its short radioactive half-life (54.5 s). Hence, a significant fraction of ^{220}Rn decays to its short-lived decay products during the time the gas remains inside the body. Because very little is known of the chemical affinity and reactivity of the ^{220}Rn, and hence, of its retention in tissue and solubility in body fluids, accurate assessment of its potential health hazard is not so well defined as for radon. However, it is commonly accepted that ^{220}Rn per se does not pose a serious health hazard under most environmental and working conditions unless inhaled in very large amounts.

The airborne decay products of ^{222}Rn and ^{220}Rn being short-lived (e.g., half-life ranging from ~3 min for ^{218}Po (RaA), to 10.6 h for ^{212}Pb (ThB)) are considerably more harmful than their radioactive gas parents. Furthermore, the ^{222}Rn and ^{220}Rn progenies are significantly more chemically and biologically active than their parents, attributes which add further to their potential health hazard. Because of their importance from an occupational standpoint, it is important to understand the complex physico-chemistry and interaction mechanisms of the short-lived decay products of ^{222}Rn and ^{220}Rn with their immediate physical environments, and biological tissue. Due to the complexity of the subject, space limitations, and for the sake of simplicity, the discussion that follows on the above topics will be a brief one.

The short-lived decay products of radon and thoron are initially formed as free atoms, usually positively charged because α-particles strip electrons away from the atom during the radioactive decay process. It is known that $\gtrsim$80% of ^{218}Po and ^{212}Pb is initially produced as single positively charged ions, whereas the remaining $\lesssim$20% occurs as neutral species (38,39). These positive ions are neutralized by recombination with negative atoms, and charge transfer, i.e., removal of electrons from neutral molecules with low ionization potentials such as NO, NO_2, and some organic vapours (40). These processes take place in times ranging from a fraction of a second to several

minutes (40).

The ions formed tend to form clusters very rapidly with water, oxygen and other trace gases in the atmosphere. Soon afterwards, and in a matter of seconds to minutes, these clusters tend to attach themselves to aerosol particles, the latter being much larger than those of the clusters. Hence, short-lived decay products of ^{222}Rn and ^{220}Rn such as ^{218}Po and ^{212}Pb are basically found in two forms:

1. Unattached, i.e., forming small clusters with trace gases present in the atmosphere; and
2. Attached to aerosols, vapours, mists, organic molecules, simple and complex atmospheric gases, etc. The physical dimensions of attached decay products are much larger than that of the unattached or cluster form.

The unattached and attached fractions, i.e., the fraction of the total in unattached and attached form, depend on a variety of factors such as:

1. Aerosol concentration, size and electrical charge distribution;
2. Chemical composition of the atmosphere;
3. Trace gas content of the atmosphere; and
4. Moisture content, temperature and other factors.

In a vacuum, or when the aerosol concentration is very low, i.e., $<<10^3$ particles cm^{-3}, the unattached fraction, f_u, can be very large, i.e., $f_u \gtrsim 0.4$ to ~1.0 (in a vacuum). For aerosol concentrations, N, greater than about 2×10^4 cm^{-3}, f_u can be considered nearly zero for all practical purposes, and the attached fraction, f_a, is therefore, very close to unity.

Because of their markedly different physical size, the diffusional properties of the attached and unattached fraction are considerably different. Although the diffusion coefficient, D, for the attached fraction varies significantly with environmental conditions and other factors (for a review of the literature see reference 41), an average value of ~0.05 cm^2 s^{-1} has been used repeatedly. For the unattached fraction, D can be from less than one to

two orders of magnitude higher than for the attached fraction.

The different diffusional properties of the attached and unattached fractions manifest themselves in the readiness for the unattached fraction to plate-out on large surfaces such as furniture, paintings, clothing, mine walls, articles of decor, and the like. These differences in the diffusivity for the two fractions is of great practical importance from the health physics standpoint. Thus, once inhaled, the attached and unattached fractions do not deposit on the same region of the respiratory system. The attached ^{222}Rn and ^{220}Rn progenies are primarily deposited in the pulmonary region, whereas the unattached fractions are deposited in the upper respiratory tract because of their higher diffusivity.

A flow chart illustrating the radon progeny pathways is shown in Figure 11. Although specifically intended for radon and its decay products, the flow chart is equally applicable to thoron and its progeny by replacing ^{222}Rn by ^{220}Rn, and so on.

A diagram that illustrates removal mechanisms for radon and its progeny is shown in Figure 12. Again, this Figure is equally applicable to thoron and its progeny. The usefulness of this illustration will become more evident when radiation control processes and methods are discussed.

Finally, this section would not be complete without some reference to radon and thoron progenies clearance rates in the respiratory system. Clearance rates are an important parameter to assess biological damage. However, most of the studies to date have been conducted using laboratory animals such as rabbits, dogs, mice and rats. Hence, the data reported in the literature should be carefully assessed when extrapolated to humans. The following data have been taken from reference 42.

Measurements of clearance rates vary widely. Pohl (cited in reference 42) found that in mice between 14 and 26% of ^{214}Pb and ^{214}Bi ions deposited in the lungs diffused away and decayed outside the lung. Morken and Scott (cited

in reference 42) estimated that in both mice and dogs most of the short-lived decay products of radon clear from lung to blood with a half-time of ~10 min. In constrast, Hurst et al., and Booker et al. (cited in reference 42) reported clearance half-times to blood in man ranging from 6.5 to 12 h for inhaled ^{212}Pb. James et al. (cited in reference 42) reports that an average 20% of ^{212}Pb was cleared with a half-time of 4 min, whereas the majority was cleared with a half-time of about 9 h.

OCCURRENCE OF RADON, THORON AND THEIR PROGENIES IN NON-URANIUM MINES

Radon and radon progeny concentration measurements have preferentially been made in uranium mines. Sparse data are available for non-uranium mines. The reason for this is that quite often it is difficult to think of uranium in non-uranium mines. Of course, the notion of absence of uranium in non-uranium mines is totally erroneous as discussed earlier. In fact, uranium minerals are often associated with other valuable mineable metal minerals. Furthermore, and more importantly, uranium, as well as thorium, are distributed more or less uniformly on the earth's crust regardless of their association with other minerals.

Thoron and thoron progeny concentration measurements have attracted serious attention only recently and in very specific cases such as in some Canadian uranium mines (e.g., in the Province of Ontario) and few other locations. No concerted, organized effort has been made at quantifying thoron and its progeny in non-uranium mines, either here in Canada or elsewhere in the world.

This section presents and reviews some of the data available regarding radon and radon progeny measurements in non-uranium mines around the world. One striking fact is worthwhile noticing, namely the lack of consistency in reporting radiation levels in non-uranium mines, a fact which adds to an already confusing state of affairs. Thus, radiation levels in non-uranium

mines are reported in either radon gas concentration levels, [^{222}Rn], or as radon progeny concentration, i.e., Working Levels (WL). (The relationship between these two variables, the so-called F-equilibrium factor, defined by F = WL/[^{222}Rn], is not a constant one and depends on a number of variables such as ventilation rate and aerosol concentration and size distribution.) Finally, a frequent omission of great practical value is the wetness condition of the mine. This condition is not normally reported and plays a very important role in the occurrence of radon and its progeny in mine openings.

As has been repeatedly indicated, uranium (and thorium) minerals are frequently 'associated' with other highly sought after mineable metal minerals such as gold, zinc, copper, cobalt and iron, among others. Furthermore, ground and surface water can dissolve large quantities of radon and transport it to distances quite far away from the source. Hence, in looking for radon in non-uranium mines, special attention should be paid to wet mines, and to some metal mines with which uranium minerals are frequently associated. It will be seen, however, that other non-metal mines, e.g., coal mines, can also exhibit high radon concentration levels. This is in agreement with the universal occurrence of uranium and thorium on earth and in natural gas, petroleum, methane, and the like. A brief review of some data available from different countries is given below.

RADON AND RADON PROGENY IN U.K. MINES

Measurements were conducted in coal, haematite, tin, fluorspar and anhydrite mines.

1. Coal Mines

Radon gas concentration measurements were made in 12 mines chosen from the East Midlands, Kent and Scottish coal fields (43). The levels found ranged from 0.6 pCiL^{-1} to 14 pCiL^{-1} with a median concentration of ~2 pCiL^{-1}.

2. Other Mines

Working Level (WL) measurements were made in six haematite (Fe_2O_3) mines, two tin mines, one fluorspar (CaF_2), and one anhydrite ($CaSO_4$) mine (44). Nearly all the measurements reported by the authors were made in those parts of the mines which were continuously occupied throughout the working week. The WL for the haematite mines were in the range 0.005 to 3.2 with a WL median ranging mostly between 0.15 to 2.0 WL. For the tin mines, WL was in the range 0.1 to 11 with a WL median of 1.1 WL and 3.7 WL for each mine. The average WL for the fluorspar mine was 0.15 whereas an average (median) of ~0.005 WL was measured at the anhydrite mine.

RADON AND RADON PROGENY IN INDIAN MINES

Radon gas concentration and WL measurements were made at four copper mines, one coal mine and one gold mine (45). The selection of the above mines among the many non-uranium mines in India was based on the number of persons employed in a particular type of mining, the depth of the mine, the nature of the ore, and the geographical distribution in the mines.

The radon gas concentrations in the gold mine (Kolar Gold Field Mines) were generally low (up to ~5 $pCiL^{-1}$, [^{218}Po] ~4 $pCiL^{-1}$) in spite of the great depths of these mines and the very long path length of the ventilation air. The author concluded that the low levels observed may be due to the absence of significant ground water movement in these mines and also the very low radium content of the ore. The radium content of the mine rock samples from different levels varied from 70 to 600 $pCi\ kg^{-1}$.

The radon levels in all the four coal mines did not show any significantly higher values than the normal atmospheric concentrations. The authors suggest that the characteristics of the coal seams and the generally very high ventilation rate maintained in the mines could be the factors responsible for the low levels observed.

All the four copper mines monitored exhibited a considerably higher

radiation level than the other mines. Working levels in excess of 0.1 were quite common, reaching values of up to 0.42. Underground water was suggested as partly responsible for these high radiation levels.

RADON AND RADON PROGENY IN U.S. COAL MINES

Radon gas concentration measurements in some U.S. coal mines (46) showed levels of up to 1.1×10^3 $pCiL^{-1}$. The highest values were obtained in West Virginia, Kentucky and Tennessee mines with an average radon gas concentration in excess of 400 $pCiL^{-1}$ (still air). Colorado and Pennsylvania coal mines followed with radon gas concentrations in the range ~0 to 147 $pCiL^{-1}$. The values quoted above are taken from several authors cited in the above reference.

RADON AND RADON PROGENY IN SCANDINAVIAN MINES

A survey of 23 non-uranium mines in Finland, Norway and Sweden was conducted (47). In 19 of the 23 mines, at least one location was found where the radon concentration was over 30 $pCiL^{-1}$. These locations were generally in unused and unventilated areas of the mine. The main source of radon was found to be water entering the mine either through exploratory drill holes or cracks in the rock. Mine water was found to contain radon in concentrations up to 3000 $pCiL^{-1}$. The greatest potential hazard was to exploratory drilling crews because of the radon entry along the drill hole. In some drilling stations, a radon concentration in excess of 1000 $pCiL^{-1}$ was observed. No information is available in the report regarding the type of mines although in one case mention is made of a lead mine which also happened to contain uranium and thorium in thukolite.

RADON AND RADON PROGENY IN CANADIAN MINES

Radon and/or radon progeny concentration measurements in non-uranium mines have been conducted in several provinces of Canada by the mining companies and/or the appropriate government regulatory agency, or other

interested agencies. However, no information, in report form or otherwise, is available to the author except for that corresponding to the provinces of Ontario, and Newfoundland and Labrador. Hence, the discussion that follows will be limited to these two Provinces.

A. Ontario Non-Uranium Mines

Sporadic measurements have been conducted for over two decades in a variety of metal mines such as gold, silver, copper, iron and nickel mines in several areas of the province including Kirkland Lake, Porcupine, Cobalt, Red Lake, Sudbury and Temagami, among others (48,49). Usually surveys have been restricted to four locations within each mine, namely, an active working place, a return air or return air circuit area, a sump pump area, and a dormant heading lacking ventilation. As expected, the highest radiation levels were, in general, found in areas with no ventilation, although exhaust and recirculation areas, and on occasion sump pump areas, also showed significant radiation levels.

Values for the radon gas concentration and radon progeny Working Level (WL) ranged from negligible to relatively high depending on the mine and the location of the mine. The higher values observed were in the following ranges:

$$WL = 0.1 \text{ to } 1.56$$

$$[^{222}Rn] = 10 \text{ to } 63 \text{ pCiL}^{-1}$$

A more detailed review of radon progeny concentrations in Ontario underground non-uranium mines has recently been presented elsewhere (50).

B. Newfoundland Non-Uranium Mines

The best documented case of high radiation levels in non-uranium mines in Canada is the fluorspar mines in St. Lawrence (Newfoundland). These mines have a history of radiation-related cancers. The mines have been extensively surveyed in the past (51,52), and more recently (53,54). Working Levels in excess of several hundred WL were observed in poorly ventilated areas. Very

high radon gas concentration levels in air and in water (in the order of many thousand $pCiL^{-1}$) have been measured in these mines. The radiation source responsible for this unusually high concentration level is radon dissolved in ground and surface water entering mine openings.

RADON AND RADON PROGENY IN OTHER NON-URANIUM MINES

Other surveys have also been conducted in countries other than those mentioned above such as Germany (55), South Africa, Czechoslovakia, Poland (55), and countries of the European Community (EC). About a decade ago, the Service de Protection Technique of the Institut de Protection et de Sûrete Nucleaire (Commissariat a l'Energie Atomique, CEA, France) was asked by the Commission of European Communities to carry out measurements to assess the collective risk to miners exposed to radon in underground non-uranium mines located in countries within the European Community (56). A survey was conducted in coal and potash mines and in a variety of metal mines (iron, blende and galena, and pyrites). The result of this survey indicates that metal mines have considerably higher radon gas concentrations (up to ~4 x 10^9 atoms m^{-3}, or 227 pCi L^{-1}) than non-metal mines (up to 2.5 x 10^8 atoms m^{-3}, or 14 pCi L^{-1}). The same survey showed the following values for the Working Level: 2 x 10^{-5} Jm^{-3} (1 WL) for metal mines and ~7.0 x 10^{-7} Jm^{-3} (0.035 WL) for non-metal mines.

To the knowledge of the author, no concerted effort has been made to gather all the available and highly scattered literature on the subject of radon and its progeny in non-uranium mines across the world in order to better assess, identify, characterize and quantify the impact on occupational health posed by the occurrence of the above radioisotopes in metal and non-metal mines.

The above information, although sketchy and incomplete, gives, nevertheless, a good idea of the potential radiation hazards in non-uranium mines that have only become more apparent in recent years as a result of more

emphasis being placed on occupational health issues and the overall well-being of the worker.

The data presented here clearly indicate that any underground non-uranium mine, if not adequately ventilated and periodically surveyed, may present a potential health problem to the working force. Almost invariably, high radiation levels are associated with wet mines. Dry mines may also present this problem when uranium minerals are found, as is frequently the case, associated with other mineable minerals. (It is interesting to notice that in some South African gold mines, uranium is obtained as a byproduct of gold.) It goes without saying that the case of wet mines with significant amounts of uranium and thorium minerals in their ores is, in general, a cause for even greater concern. Reducing or controlling radon, thoron, and their progeny in non-uranium mines is necessary and this is the subject of the brief review that follows.

CONTROL OF RADIATION IN NON-URANIUM MINES

The methods available for reducing or controlling radiation levels in non-uranium mines are, with rare exceptions, similar to those used in uranium mines. The most widely used method is forced mechanical ventilation which operates by dilution of airborne pollutants in mine air, such as diesel particulates, respirable dust, noxious gases, organic vapours, mists, and naturally, radon, thoron, and their decay products. A ventilation modality which is receiving wide attention recently on account of its economical advantages is controlled recirculation. However, there are other methods and techniques that could be effective and useful, notwithstanding economic considerations. Some of these methods are listed below:

1. Mechanical filtration with conventional filters or using special filters with electret-like characteristics for more efficient particulate removal;
2. Removal of pollutants by using activated charcoal beds;

3. Combination of items 1 and 2;
4. Water scrubbing;
5. Electrostatic precipitation;
6. Removal of radioactive aerosols by negative-ion generators;
7. Plate-out of radioactive aerosols by electrostatic screens;
8. Deflection by air curtains;
9. Coating of mine walls with radon 'barriers'; and
10. Dewatering or drainage of mine water.

The methods indicated in items 1, and 5 to 7 are effective, to varying degrees, in the removal of radon and thoron decay products from mine air. Although of great value, this does not entirely solve the radiation problem because the radioactive gases ^{222}Rn and ^{220}Rn are totally unaffected, and hence, will continue to decay into their short-lived decay products.

The only effective method to remove airborne radon and thoron is by using beds of activated charcoal (see item 2). Item 9 is partially successful, albeit quite expensive, and full of practical difficulties when applied to prevent radon diffusion or transport through mine walls. Method 10 is efficient when radon is dissolved in ground and surface water reaching mine openings. Method 4, i.e., water scrubbing is quite efficient in removing long-lived radioactive dust (or 'normal' dust) in the respirable size range, preferably for particles of size $\gtrsim 3$ μm. The method is inefficient in the removal of radioactive aerosols in the submicron size range. Method 8 is not effective for radon or thoron, but may somewhat reduce the concentration level of their decay products by plate-out on mine walls caused by air turbulence and convective transport.

CONCLUSIONS

Uranium and thorium minerals are widely distributed in the earth's crust. The radioactive decay of these radionuclides leads to the formation of

two radioactive gases (and their decay products), namely, radon (^{222}Rn) and thoron (^{220}Rn), respectively, which readily find their way through interstitial spaces, and along cracks, fissures and faults in the rock formation and ground by diffusion and convective transport mechanisms.

Because of its chemical inertness, high diffusivity (aided by air and water transport mechanisms), and its relatively long radioactive half-life, radon finds its way into living spaces and working environments such as homes and non-uranium mines. Theoretical principles and experimental evidence show that elevated levels of radon and its short-lived decay products can occur in non-uranium mines if not surveyed and adequately ventilated, thereby posing a potential occupational health hazard.

REFERENCES

1. The Hazards to Man of Nuclear and Allied Radiations. A second report to the Medical Research Council, Medical Research Council, London (U.K.); December 1960.

2. Lindel, B. and Dobson, L.R., "Ionizing radiation and health"; Public Health Papers No. 6, World Health Organization, Geneva; 1961.

3. Sources and Effects of Ionizing Radiation. United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR); 1977 Report, United Nations, New York; 1977.

4. The Natural Radiation Environment. J.A.S. Adams and W.M. Lowder (Eds.), University of Chicago Press, Chicago; 1964.

5. Cameron, I.R., "Meteorites and cosmic radiation"; Scientific American, vol. 229, No. 1, pp. 65-73; 1973.

6. Cachon, A., Daudin, A. and Jauneau, L., "Cosmic rays"; Walker and Company, New York; 1965.

7. Hooper, J.E. and Scharff, M., "The cosmic radiation"; Methuen and Co. Ltd., London, and John Wiley and Sons Inc., New York; 1958.

8. Erläuterungeu zur Nuklidkarte (2. Auflage, Nachdruck 1963), Kernforschungszentrum (Karsruhe, FRG), Institut für Radiochemie, 1963.

9. Jastrow, R. and Thompson, M.H., "Astronomy: fundamentals and frontiers"; John Wiley and Sons Inc., New York/London; 1975.

10. Mason, B., "Principles of geochemistry"; John Wiley and Sons Ltd., New York, NY, 3rd edition; 1966.

11. Tatsch, J.H., "Uranium deposits: origin, evolution, and present characteristics"; Tatsch Associates, Sudbury, Massachusetts 01776; 1976.

12. The Theoretical and Practical Aspects of Uranium Geology; The Royal Society (London); 1979.

13. Stein, L., "Chemical properties of radon"; in Radon and Its Decay Products - Occurrence, Properties, and Health Effects; P.K. Hopke (Ed.); pp. 240-251; American Chemical Society, Washington, DC; 1987.

14. Bartlett, N. and Sladky, F.O., "The chemistry of krypton, xenon and radon"; in: Comprehensive Inorganic Chemistry; A.F. Trotman-Dickenson (Ed.); vol. 1, pp. 213-330; Pergamon Press, Oxford, U.K.; 1973.

15. Avronin, V.V., Krasikova, R.N., Nefedov, V.D. and Toropova, M.A., "Coprecipitation of radon oxide with cesium fluoroxenate"; Radiokhim., vol. 27, pp. 511-514; 1985.

16. Tanner, A.B., "Geological factors that influence radon availability"; in: Indoor Radon. Proc. of APCA Int. Specialty Conf. (TT-7 Indoor Air Quality Committee), Philadelphia, PA; Feb. 1986.

17. Rutherford, E., Chadwick, J. and Ellis, C.D., Radiation from radioactive substances, Cambridge University Press, Cambridge; 1930.

18. Makower, W. and Geiger, H., Practical measurements in radio-activity. Longmans, Green and Co., London; 1912.

19. Lind, S.C. (with Hochanadel, C.J. and Ghormley, J.A.), Radiation chemistry of gases. (American Chemical Society Monograph Series); Reinhold Publishing Corp., New York.

20. Tanner, A.B., "Radon migration in the ground: a supplementary review"; in: Natural Radiation Environment III. T.F. Gesell and W.M. Lowder (Eds.); CONF-780422, vol. 1, pp. 5-56; Springfield, VA; NTIS; 1980.

21. Fleischer, R.L., "Moisture and ^{222}Rn emanation"; Health Phys., vol. 52, pp. 797-799; 1987.

22. Greiner, N.R., "Radon emanation from coals: effects of moisture and particle size"; Health Phys., vol. 48, pp. 283-288; 1985.

23. Harvey, B.G., "Recoil techniques in nuclear reaction and fission studies"; Ann. Rev. Nuclear Sci., vol. 10, p. 235; 1960.

24. Tanner, A.B., "Radon migration in the ground"; in: The Natural Radiation Environment. J.A.S. Adams and W.M. Lowder (Eds.), pp. 161-190; The University of Chicago Press, Chicago; 1964.

25. Stranden, E., Kolstad, A.K., Lind, B., "Radon exhalation: moisture and temperature dependence"; Health Phys., vol. 47, pp. 480-484; 1984.

26. Strong, K.P. and Levins, D.M., "Effect of moisture content on radon emanation from uranium ore and tailings"; Health Phys., vol. 42, pp. 27-32; 1982.

27. Fukui, M., "Soil water effects on concentration profiles and variations of ^{222}Rn in a vadose zone"; Health Phys., vol. 53, pp. 181-186; 1987.

28. Megumi, K., and Mamuro, T., "Emanation and exhalation of radon and thoron from soil particles"; J. Geophys. Res., vol. 79, pp. 3357-3360; 1974.

29. Evans, R.D., "Engineer's guide to the elementary behaviour of radon daughters"; Health Phys., vol. 17, p. 229; 1969.

30. Schroeder, G.L. and Evans, R.D., "Some basic concepts in uranium mine ventilation"; Trans SME-AIME 244, p. 301; 1969.

31. Thomas, J.W. and Epps, J.W., "Radium A, B, and C concentrations and Working Levels from radon emanation in a mine tunnel"; HASL-TM 70-4; (available from U.S. Dept. of Energy, NTIF, P.O. Box 62, Oak Ridge, TN 37830); 1970.

32. Rolle, R., "Radon daughters and age of ventilation air"; Health Phys., vol. 23, p. 118; 1972.

33. Beckman, R.T. and Holub, R.F., "Radon daughter growth with continuous radon influx and various ventilation rates"; MSHA Investigation Report IR-1099; U.S. Dept. of Labor, Washington, DC; 1979.

34. Bigu, J., "Theoretical models for determining ^{222}Rn and ^{220}Rn progeny levels in Canadian underground U mines - a comparison with experimental data"; Health Phys., vol. 48, pp. 371-399; 1985. (See also CANMET Division Report MRP/MRL 82-149(J); 1982.)

35. Bigu, J., "Non steady-state radiation mine models - practical applications to personal dosimetry and environmental monitoring"; Proc. Int. Conf. Occup. Radiation and Safety in Mining, Toronto, October 1984. Canadian Nuclear Association; H. Stocker (Ed.), Toronto, pp. 429-439; 1985.

36. Bigu, J., "The effect of time-dependent ventilation rates in partially enclosed radioactive environments"; Proc. 1st Int. Symp. Ventilation for Contaminant Control (Ventilation'85, Toronto, October 1985). Elsevier, Amsterdam; E.D. Goodfellow (Ed.); pp. 477-488; 1985.

37 Bigu, J., "The effect of time-dependent ventilation and radon (thoron) gas emanation rates in underground uranium mines"; Proc. 3rd Mine Ventilation Symp.; J.M. Mutmansky (Ed.); Chapter 52, pp. 353-362; Society of Mining Engineers Inc., Littleton, CO; 1987.

38. Porstendörfer, J. and Mercer, T.T., "Influence of electric charge and humidity upon the diffusion coefficient of radon decay products"; Health Phys. vol. 37, pp. 191-199; 1979.

39. Dua, S.K., Kotrappa, P. and Gupta, P.C., "Influence of relative humidity on the charged fraction of decay products of radon and thoron"; Health Phys., vol. 45, pp. 152-156; 1983.

40 Raes, F., "Description of the properties of unattached Po-218 and Pb-212 particles by means of the classical theory of cluster formation"; Health Phys., vol. 49, pp. 1177-1187; 1985.

41. Scofield, P., "Radon decay product in-door behaviour - parameter, measurement method, and model review"; Report SSI 88-07; National Institute of Radiation Protection, Stockholm; 1988.

42. Greenhalgh, J.R., James, A.C. and Smith, H., "Clearance of radon daughters from the lung"; Ann. Research and Development Report 1976; National Radiological Protection Board (NRPB), pp. 49-51; 1977.

43. Duggan, M.J., Howell, D.M. and Soilleux, P.J., "Concentration of radon-222 in coal mines in England and Scotland"; Nature (London), vol. 219, p. 1149; 1968.

44. Duggan, M.J., Soilleux, P.J., Strong, J.C. and Howell, D.M., "The exposure of United Kingdom miners to radon"; Brit. J. Ind. Med., vol. 27, pp. 106-109; 1970.

45. Nair, N.B., Eapen, C.D. and Rangarajan, C., "High radon levels in some non-uranium mines in India"; in: Natural Radiation Environment; Vohra, Pillai, Mishra and Sadasivan (Eds.); John Wiley and Sons, New York; 1982.

46. Gogolak, C.V., "Review of ^{222}Rn in natural gas produced from unconventional sources"; Report DOE/EML-385; U.S. Dept. of Energy, Environmental Measurements Laboratory, New York, NY; 1980.

47. Kahlos, H., "Radonongelma suomen kaivoksissa"; translated into English by R. Tervo under the title 'Radon problem in Finnish mines'; Division Report MRP/MRL 77-102(T); CANMET, Energy, Mines and Resources Canada; Sept. 1977.

48. Seguin, H., "Radiation health effects and W.C.B. criteria"; presented to Industrial Disease Standards Panel of the Ontario Ministry of Labour, Toronto, Ont.; Sept. 1986.

49. Coughlan, W.K., Private communication; October 1988.

50. Vergunst, J., "Development of Ontario's proposed radon daughter regulation for non-uranium mines"; In: Radiation Measurement and Control in Non-Uranium Mines. (Seminar organized by the Resource Technology Centre and CAIRS, Nov. 28-29, Elliot Lake, Ontario, 1988).

51. Yourt, G.R., "Radiation and ventilation underground at Newfoundland Fluorspar Works"; Report of Investigation; G. Reub Yourt, Consultant Engineer, Toronto, Canada; 1974.

52. Corkill, D.A. and Dory, A.B., "A retrospective study of radon daughter concentrations in the work place in the Fluorspar Mines of St. Lawrence, Nfld"; Information Report AECB INFO-0127; Atomic Energy Control Board; July 1984.

53. Bigu, J., Frattini, A. and Edwardson, E., "Radioactivity conditions in a non-uranium underground mine in the absence of mining operations"; Division Report MRL 88-67(TR); CANMET, Energy, Mines and Resources Canada; May 1988.

54. Bigu, J., Edwardson, E. and Frattini, A., "Radioactivity conditions in a non-uranium underground mine during mining operations"; Division Report MRL 88-94(TR); CANMET, Energy, Mines and Resources Canada; July 1988.

55. In Proc. Radiation Hazards in Mining: Control, Measurement and Medical aspects. M. Gomez (Ed.); pp 629-641, SME/AIMMPE, New York, NY, 1981.

56. Bernhard, S., Le Gac, J.A., Seguin, H., Zeetwoog, P.R., "Radon levels and radon daughter exposures of workers in non-uranium mines of the E.C."; in Proc. Radiation Hazards in Mining: Control, Measurement and Medical Aspects; M. Gomez (Ed.); pp. 625-628, SME/AIMMPE, New York, NY; 1981.

Table 1 - Naturally occurring radionuclides with half-lives greater than 10^8 years (y).

Radionuclide	Z	Radioactive Decay Mode	Radioactive Half-life (y)
^{40}K	19	β, K, γ	1.27×10^9
^{50}V	23	K	4.8×10^{14}
^{87}Rb	37	β	4.7×10^{10}
^{115}In	49	β	6×10^{14}
^{138}La	57	β, K	1.1×10^{11}
^{144}Nd	60	α	2.4×10^{15}
^{147}Sm	62	α	1.2×10^{11}
^{152}Gd	64	α	1.1×10^{14}
^{176}Lu	71	β, γ	2.2×10^{10}
^{174}Hf	72	α	2×10^{15}
^{187}Re	75	β	6×10^{10}
^{190}Pt	78	α	7×10^{11}
^{192}Pt	78	α	$\sim 10^{15}$
^{204}Pb	82	α	1.4×10^{17}
^{232}Th	90	α, γ	1.39×10^{10}
^{235}U	92	α, γ, sf	7.1×10^8
^{238}U	92	α, γ, sf	4.5×10^9

Notes: α, β and γ indicate radioactive decay mode by α-particle, β-particle, and γ-ray emission, respectively.

The symbol sf if used to indicate spontaneous fission.

The letter K indicates radioactive decay by K-electron capture.

Table 2 - Abundance of some elements in the earth's crust.

Element	Weight Percent	Volume Percent
O	46.60	93.77
Si	27.72	0.86
Al	8.13	0.47
Fe	5.00	0.43
Ca	3.63	1.03
Na	2.83	1.32
K	2.59	1.83
Mg	2.09	0.29
Ti	0.44	-
H	0.14	-
P	0.12	-
Total	99.29	~100

Table 3 - Properties of some relevant physical elements.

Element	Z	AW	MP °C	BP °C	ρ	IP eV	Valence	$T_{1/2}$
U	92	238.03	1132.3	3818	18.95	-	3,4,5,6	4.51×10^9 y
Th	90	232.04	~1700.0	~4000	~11.66	5.7	4	1.39×10^{10} y
Ra	88	226	700	<1737	5 ?	-	2	1620 y
Rn	86	222	-71	-61.8	4.4*	10.74	0^+	3.82 d
Rn	86	220	-	-	-	-	0 ?	54.5 s
Po	84	~210	252	960	9.42	-	2,4,6	-
Bi	83	208.98	271.3	1562	9.75	9.0	3,5	-
Pb	82	207.19	327.5	1740	11.35	7.41	1,2,4	-

Notes: The symbols Z, AW, MP, BP, ρ, IP, and $T_{1/2}$ stand, respectively, for atomic number, atomic weight, melting point, boiling point, specific gravity, ionization potential, and radioactive half-life.

* The density of ^{222}Rn is 9.72 g/L (°C, 1 atm.). The specific gravity (ρ) given above refers to its liquid form at -62°C. The gas liquifies at -65°C and solidifies at -100°C.

The properties for Po, Bi, and Pb tabulated above correspond to the stable isotopes. The chemical and physical properties of their respective radioisotopes may differ considerably from that of the stable isotopes.

The properties tabulated above for U and Th correspond to the naturally occurring isotopic mixtures. Their half-lives correspond to ^{238}U and ^{232}Th, respectively.

The radioactive half-lives of the short-lived decay products of ^{222}Rn and ^{220}Rn, i.e., radioisotopes of Po, Bi, and Pb are shown in Figures 1 to 3.

\+ Radon has recently been shown to exist in several chemical compounds (artificially produced) in a +2 oxidation state.

Table 4 - Radium and thorium concentration in coal and coal residues.

Coal or Coal Residue	Location	$[^{226}Ra]$ (pCi g^{-1})	$[^{228}Th]$ (pCi g^{-1})	[U] ($\mu g\ g^{-1}$)
Coal	Australia	0.8-1.3	-	
Coal	Czechoslovakia	0.11-0.35	-	
Coal	FRG	<0.7	<0.6	
Bituminous coal	Hungary	0.04	-	
Bituminous coal	Poland	0.048-0.94	-	
Coal	U.S.A.	0.014-0.6	0.02-0.07	
Coal ash	Australia	4.7-8.3		
Coal ash	FRG	6.2	2.6	
Coal ash	Japan	0.1-0.98	2.35-15.3	
Coal ash (semi-bituminous)	U.S.A.	3.8	2.6	
Slag	Poland	4.3	1.2	
Slag	U.S.A.	0.55-4.5	0.5	
Fly ash	Australia	14.0	-	
Fly ash	Hungary	0.6-15		
Fly ash	Poland	0.61-6.4	1.1	
Fly ash	U.S.A.	0.4-3.1	0.4	
Anthracite (coal)	U.S.A.	-	-	1.5*
Anthracite (ash)	U.S.A.	-	-	9.0*
Eastern bituminous (coal)	U.S.A.	-	-	1.9*
Eastern bituminous (ash)	U.S.A.	-	-	38*
Western bituminous (coal)	U.S.A.	-	-	1.9*
Western bituminous (ash)	U.S.A.	-	-	16*
Lignite (coal)	U.S.A.	-	-	2.3*
Lignite (ash)	U.S.A.	-	-	22*

* Values taken from NCRP Report No. 77 (National Council on Radiation Protection and Measurements, 1984). The rest of the data taken from Sources and Effects of Ionizing Radiation (UNSCEAR, 1977 Report, United Nations, New York, 1977).

Table 5 - Average radon gas concentration in natural gas at the well and in the distribution line.

Location		$[^{222}Rn]_{ave.}$ (pCi L^{-1})	$[^{222}Rn]^{+}$ (pCi L^{-1})	Remarks
Borneo		-	1.5-3.2	Well
Canada	Alberta	62	-	Well
	B.C.	473	-	Well
	Ontario	167	-	Well
FRG		-	1.0-9.6	Well
Netherlands		-	1.1-44.7	Well
Nigeria		-	0.9-2.9	Well
North Sea		1.3	-	Well
U.S.A.		25-100*	-	Well
Poland		8	-	Distribution line
U.S.A.		1.5-50.5*	-	Distribution Line

* Range of average values corresponding to two different states.

+ Range of values quoted when average values are not available.

The square brackets are used to denote concentration. The index ave. indicates average.

Note: The data given above have been taken from 'Sources and Effects of Ionizing Radiation' (UNSCEAR Report 1977, United Nations, New York, 1977).

Table 6 - Radon and thoron data in several materials*.

Gas	Medium	Porosity	Diffusion Coefficient ($cm^2\ s^{-1}$)	Diffusion* Length (cm)	Remarks
Radon	Dry silt, sand and gravel	0.21-0.53	$5.4\text{-}7.6\times10^{-2}$	161-190	
	Water saturated clay	0.27-0.71	$1.2\text{-}4.5\times10^{-6}$	0.7-1.47	
	Mud (1.57 gm/cm^3)		5.7×10^{-6}		Air, 37.2% moisture, 19-20°C
	Mud (1.02 gm/cm^3)		2.2×10^{-6}		Air, 85.5% moisture, 19-20°C
	Rock	0.062	2.0×10^{-3}	124	
	Rock	0.25	3.0×10^{-2}	239	
	Dense rock		0.5×10^{-3}		
	Sandstone	0.50	2.2×10^{-2}	145	
	Unconsolidated glacial debris (Lincoln, Mass)	0.40	2.0×10^{-2}	154	
	Consolidated sandstone (Grants, New Mexico)	0.25	3.0×10^{-2}	239	
	Alluvium (Yucca Flat)	Dry, sandy	3.6×10^{-2}		
	Alluvium (Yucca Flat)	0.25	0.10	436	Very dry, powdery, sparse to no ground cover
	Concrete	0.05	1.69×10^{-5}	12.7	
	Concrete	0.25	3.08×10^{-5}	7.6	
	Shotcrete	0.05	1.69×10^{-5}		
	Uranium tailings	0.54	0.80×10^{-3}	26.6	
	Uranium tailings	0.40	1.20×10^{-3}	37.8	10% moisture
	Uranium tailings		1.70×10^{-3}		5% moisture
	Uranium tailings		0.20×10^{-3}		15% moisture
Thoron	Building sand (1.4 gm/cm^3)	0.39	5.4×10^{-2}	3.3	Air, 4% moisture
	Fine sand (mostly quartz)		6.8×10^{-2}		Air, 0% moisture
	Fine sand (mostly quartz)		5.0×10^{-3}		Air, 17% moisture

*Data taken from Sogaard-Hansen, J. and Damkjaer, A., Health Phys. vol. 53, pp. 455-459, 1987; Schroeder et al., J. Geophys. Res. vol. 70, pp. 471-474; and Tanner, A.B. (and refs. therein) in The Natural Radiation Environment, J.A.S. Adams and W.M. Lowder (eds.), pp. 161-190, The University of Chicago Press, Chicago, 1964.

THE ACTINIUM SERIES

Radioelement	Corresponding element	Symbol	Radiation	Half-life
Actinouranium	Uranium	^{235}U	α	7.07×10^{8} yr
↓				
Uranium Y	Thorium	^{231}Th	β	25.6 hr
↓				
Protactinium	Protactinium	^{231}Pa	α	3.25×10^{4} yr
↓				
Actinium	Actinium	^{227}Ac	β and α	21.7 yr
98.8% | 1.2%				
↓ | ↓				
Radioactinium	Thorium	^{227}Th	α	18.2 days
Actinium K	Francium	^{223}Fr	β	21 min
Actinium X	Radium	^{223}Ra	α	11.7 days
Ac Emanation	Radon	^{219}Rn	α	3.92 sec
Actinium A	Polonium	^{215}Po	α and β	1.83×10^{-3} sec
~ 100% | ~ 5×10^{-4}%				
↓ |				
Actinium B	Lead	^{211}Pb	β	36.1 min
Astatine-215	Astatine	^{215}At	α	~ 10^{-4} sec
↓				
Actinium C	Bismuth	^{211}Bi	β and α	2.16 min
99.69% | 0.32%				
↓ |				
Actinium C'	Polonium	^{211}Po	α	0.52 sec
Actinium C''	Thallium	^{207}Tl	β	4.76 min
↓				
Actinium D (end product)	Lead	^{207}Pb	None	Stable

Fig. 1 - The actinium series.

THE URANIUM SERIES

RADIOELEMENT	CORRESPONDING ELEMENT (2)	SYMBOL	RADIATION	HALF-LIFE
Uranium I	Uranium (92)	^{238}U	α	4.51×10^9 yr
↓				
Uranium X_1	Thorium (90)	^{234}Th	β	24.1 days
↓				
Uranium X_2^*	Protactinium (91)	^{234}Pa	β and I.T.	1.17 min
99.87% | 0.13%				
↓				
Uranium II	Uranium (92)	^{234}U	α	2.48×10^5 yr
Uranium Z	Protactinium (91)	^{234}Pa	β	6.66 hr
Ionium	Thorium (90)	^{230}Th	α	7.5×10^4 yr
↓				
Radium	Radium (88)	^{226}Ra	α	1.62×10^3 yr
↓				
Ra Emanation	Radon (86)	^{222}Rn	α	3.82 days
↓				
Radium A	Polonium (84)	^{218}Po	α and β	3.05 min
99.96% | 0.04%				
↓				
Radium B	Lead (82)	^{214}Pb	β	26.8 min
Astatine-218	Astatine (85)	^{218}At	α	2 sec
↓				
Radium C	Bismuth (83)	^{214}Bi	β and α	19.7 min
99.96% | 0.04%				
↓				
Radium C'	Polonium (84)	^{214}Po	α	1.5×10^{-4} sec
Radium C''	Thallium (81)	^{210}Tl	β	1.32 min
↓				
Radium D	Lead (82)	^{210}Pb	β	19.4 yr
↓				
Radium E	Bismuth (83)	^{210}Bi	β and α	2.6×10^6 yr
~ 100% | ~ 10^{-5}%				
↓				
Radium F	Polonium (84)	^{210}Po	α	138.4 days
Thallium-206	Thallium (81)	^{206}Tl	β	4.23 min
↓				
Radium G (end product)	Lead (82)	^{206}Pb	None	Stable

* Undergoes isomeric transition (I.T.) to form uranium Z (^{234}Pa); the latter has a half life of 6.66 hr. emitting β radiation and forming Uranium II ^{234}U.

Fig. 2 - The uranium series.

THE THORIUM SERIES

RADIOELEMENT	CORRESPONDING ELEMENT	SYMBOL	RADIATION	HALF-LIFE
Thorium	Thorium	^{232}Th	α	1.39×10^{10} yr
↓				
Mesothorium I	Radium	^{228}Ra	β	6.7 yr
↓				
Mesothorium II	Actinium	^{228}Ac	β	6.13 hr
↓				
Radiothorium	Thorium	^{228}Th	α	1.90 yr
↓				
Thorium X	Radium	^{224}Ra	α	3.64 days
↓				
Th Emanation	Radon	^{220}Rn	α	54.5 sec
↓				
Thorium A	Polonium	^{216}Po	α	0.16 sec
~ 100% | 0.014%				
↓				
Thorium B	Lead	^{212}Pb	β and α	10.6 hr
Astatine-216	Astatine	^{216}At	α	3×10^{-4} sec
↓				
Thorium C	Bismuth	^{212}Bi	β and α	60.5 min
66.3% | 33.7%				
↓				
Thorium C'	Polonium	^{212}Po	α	3×10^{-7} sec
Thorium C''	Thallium	^{208}Tl	β	3.1 min
↓				
Thorium D (end product)	Lead	^{208}Pb	None	Stable

Fig. 3 - The thorium series.

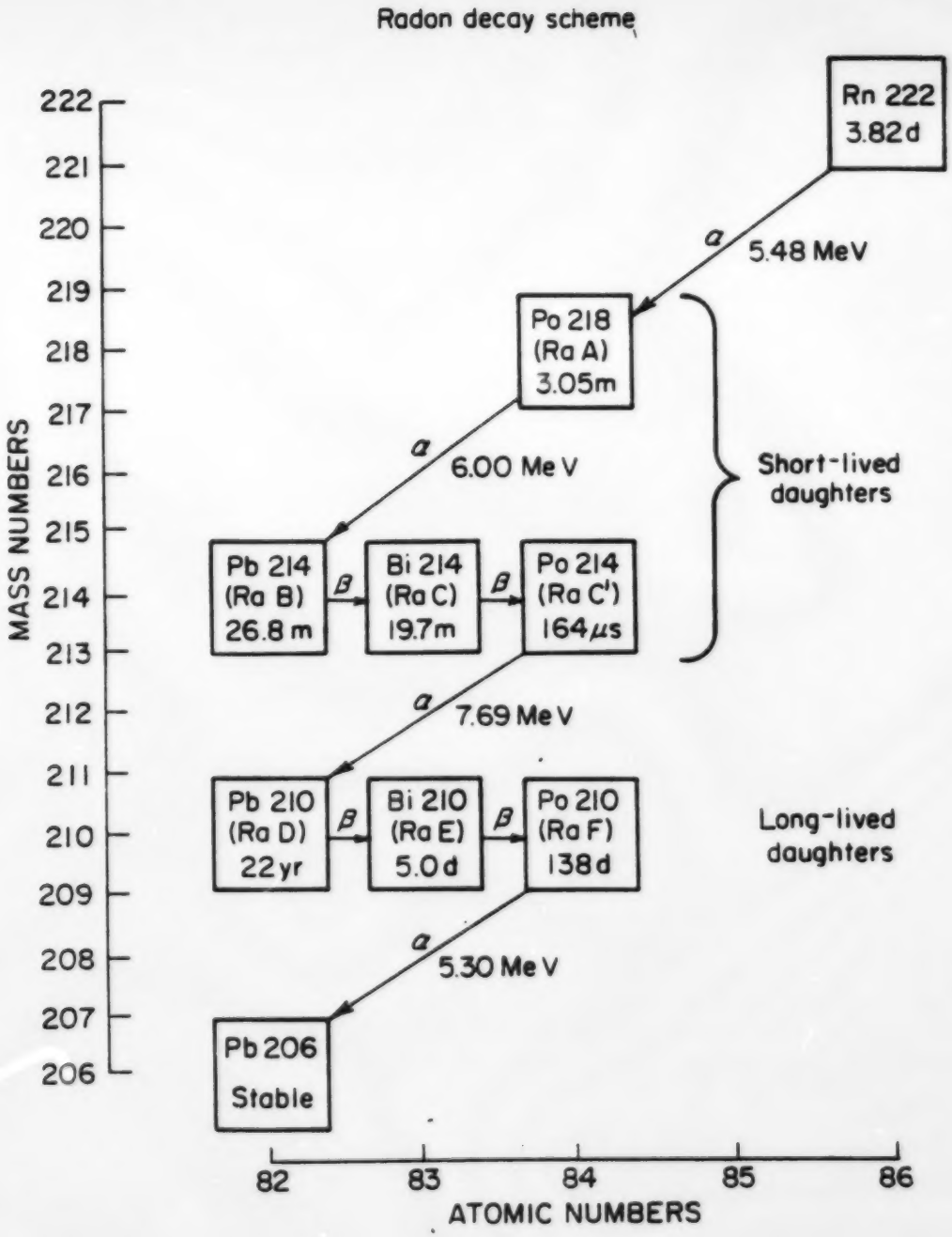

Fig. 4 - The radon decay scheme.

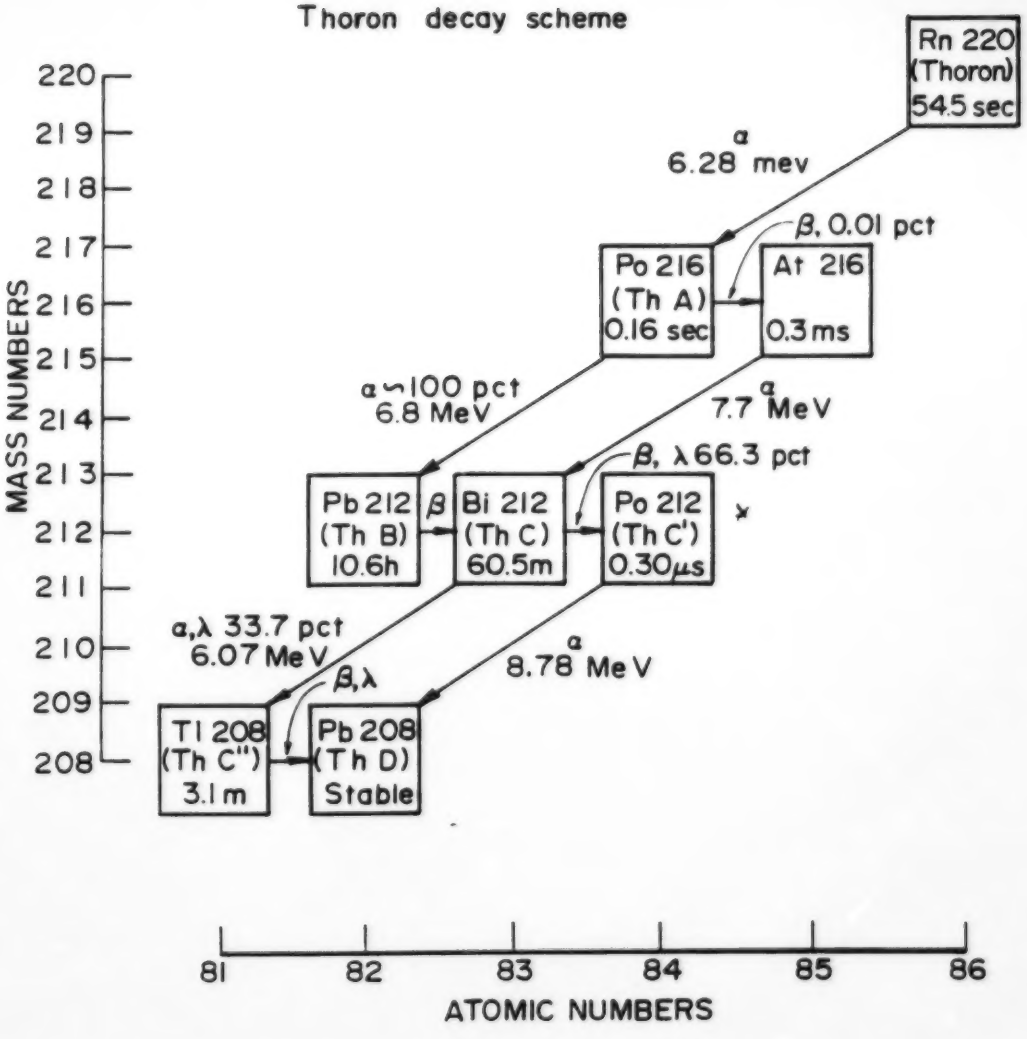

Fig. 5 - The thoron decay scheme.

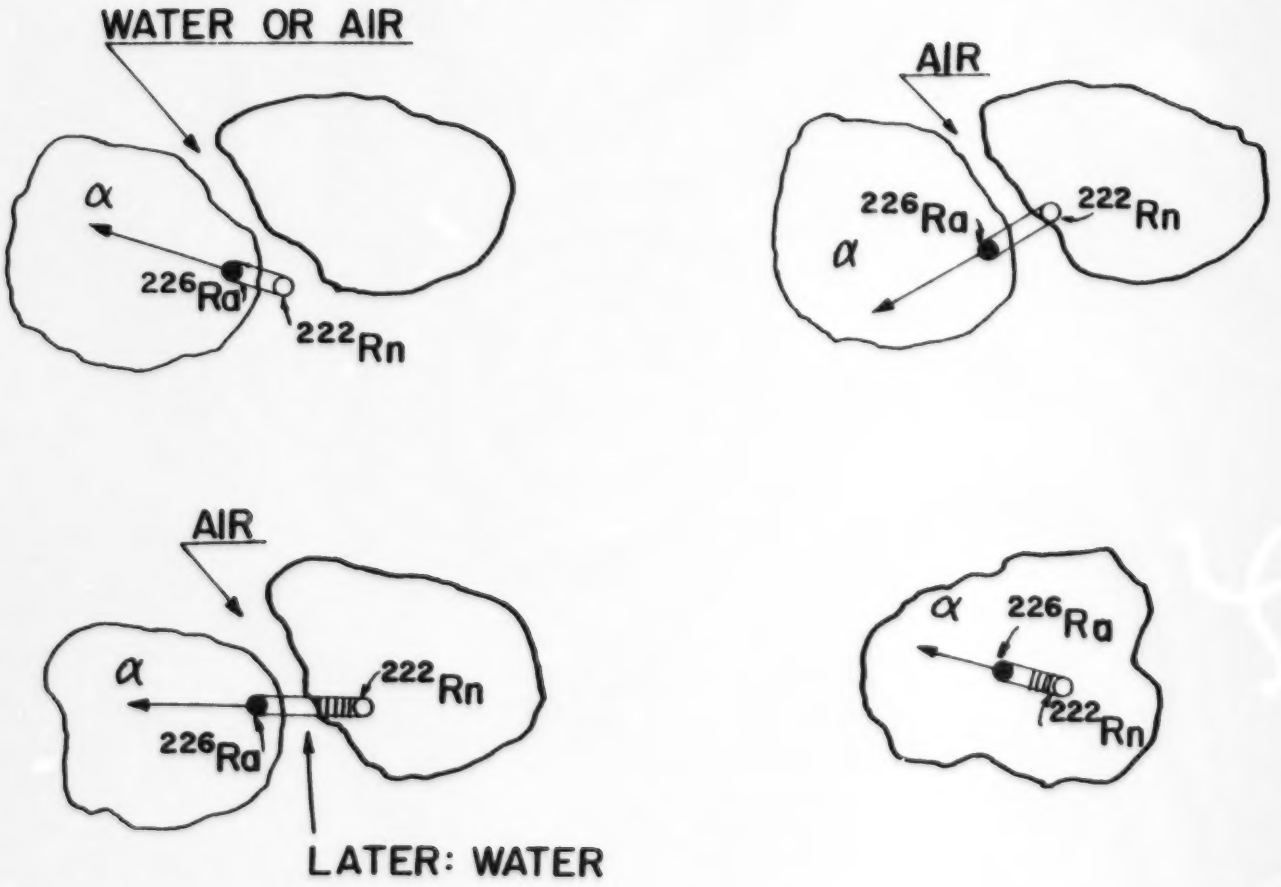

Fig. 6 - Mechanisms of ^{222}Rn release from solids.

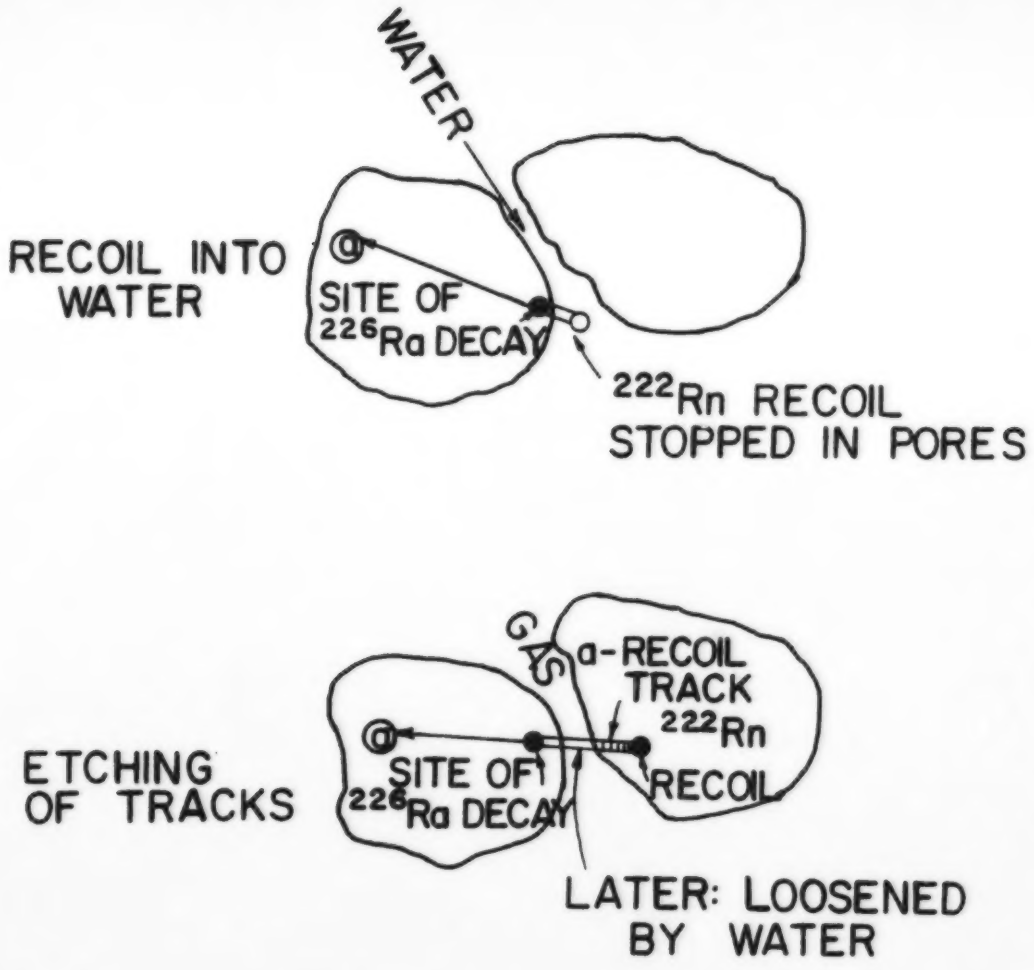

Fig. 7 - Mechanisms of ^{222}Rn release from solids.

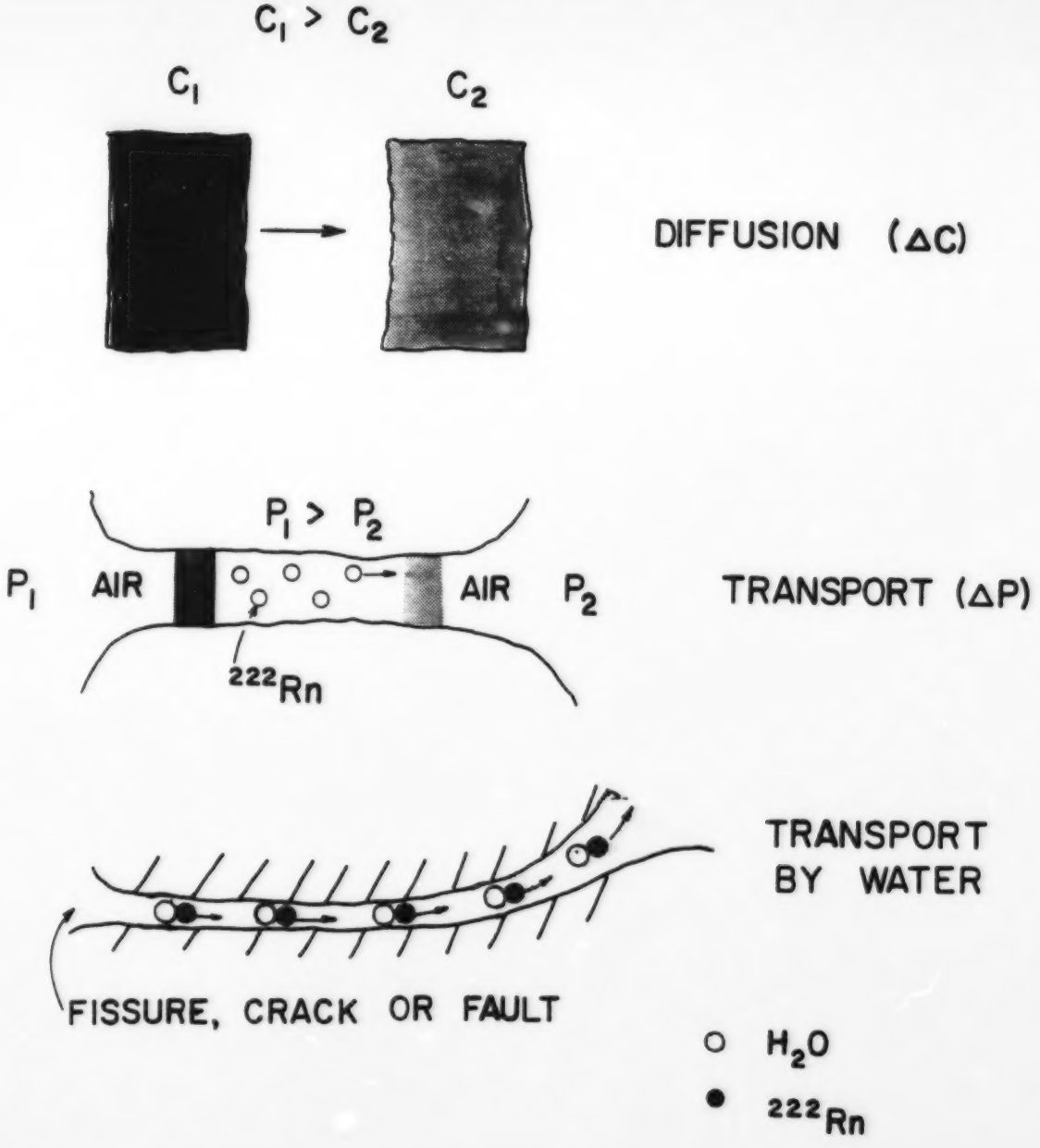

Fig. 8 - Mechanism of ^{222}Rn and ^{220}Rn escape from soils or fragmented solids.

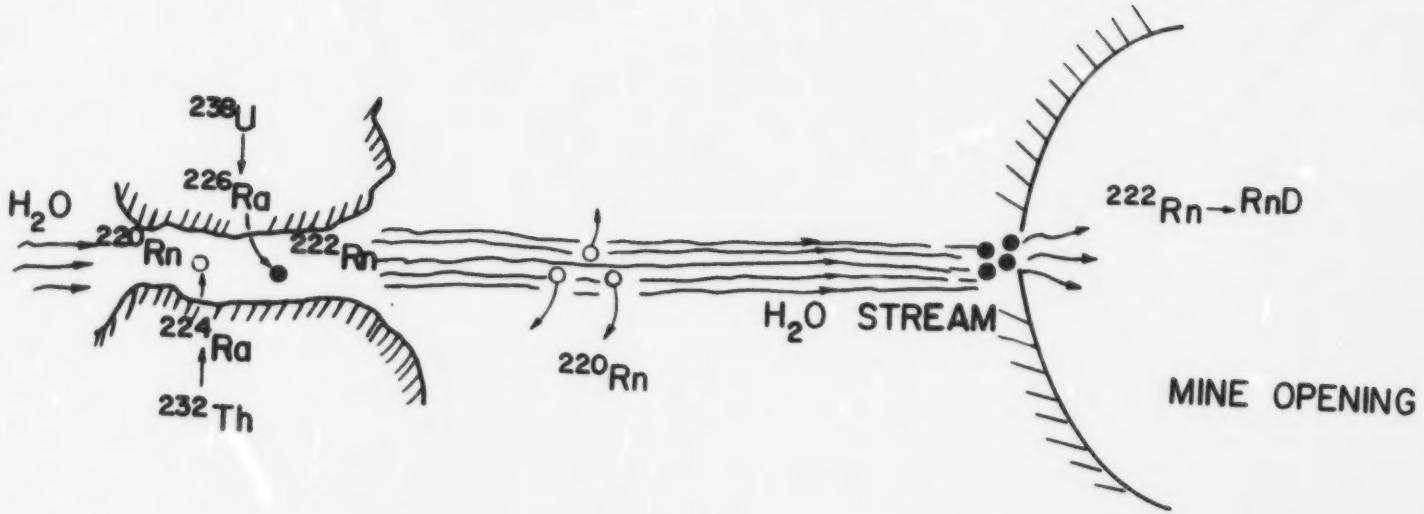

Fig. 9 - Mine with no U, Th, or Ra content in walls, but with water entry.

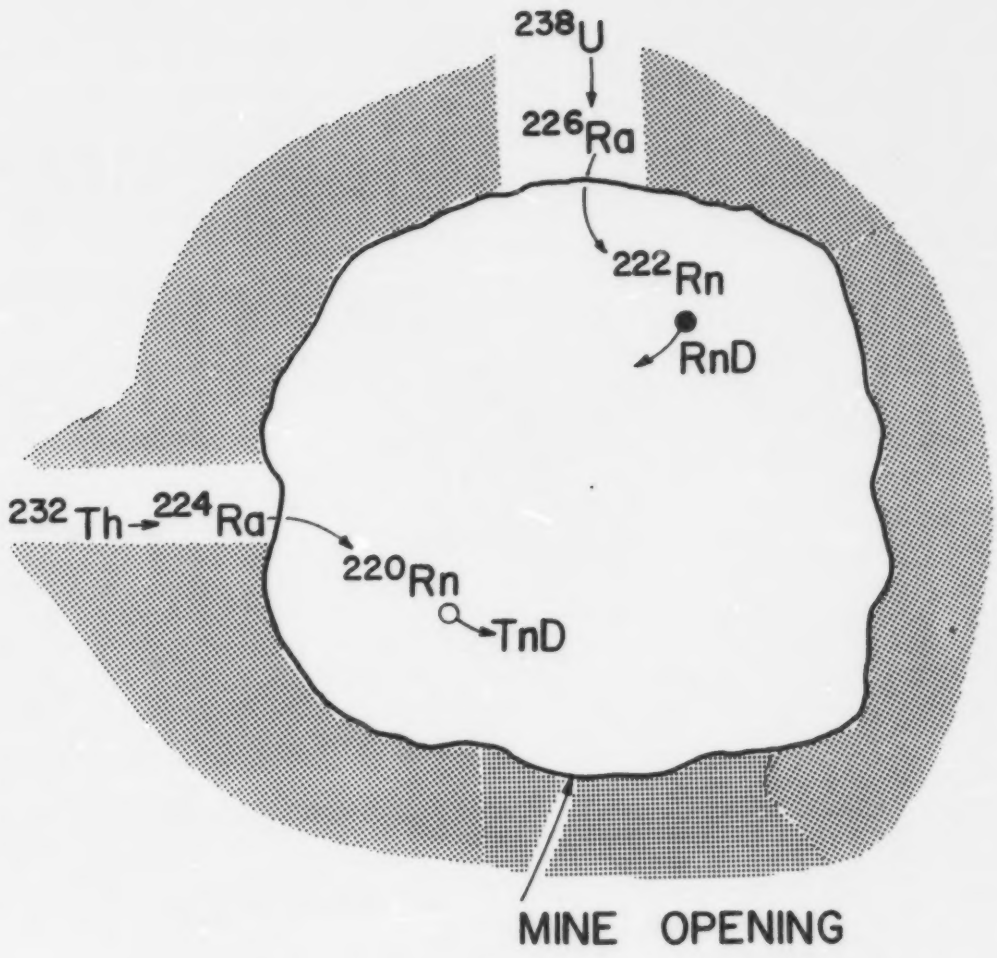

Fig. 10 - Mine with appreciable amounts of ^{238}U, ^{226}Ra, ^{232}Th, and ^{224}Ra, but no water entry.

FLOW CHART OF INDOOR RADON PROGENY PATHWAYS

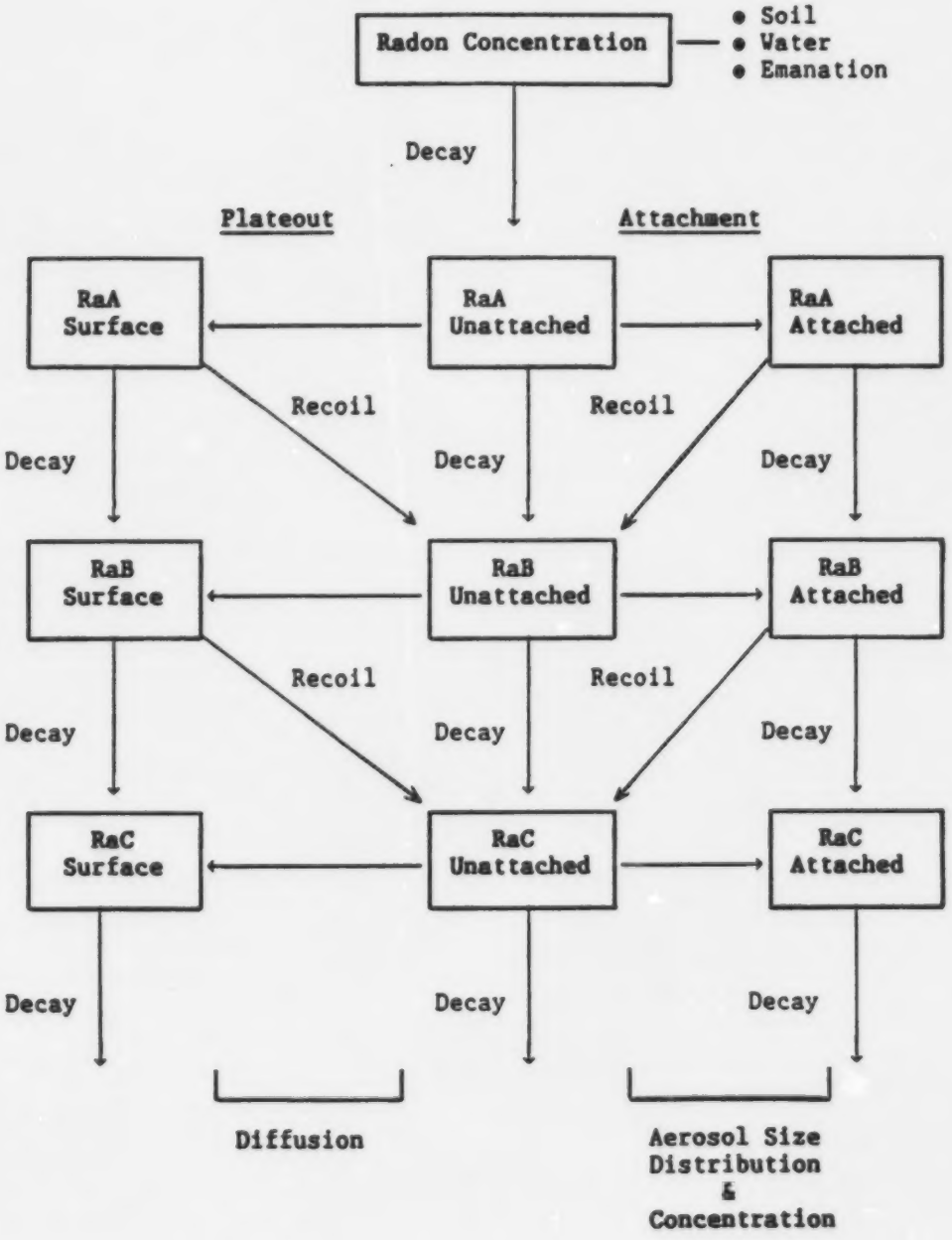

Fig. 11 - Radon progeny pathways.

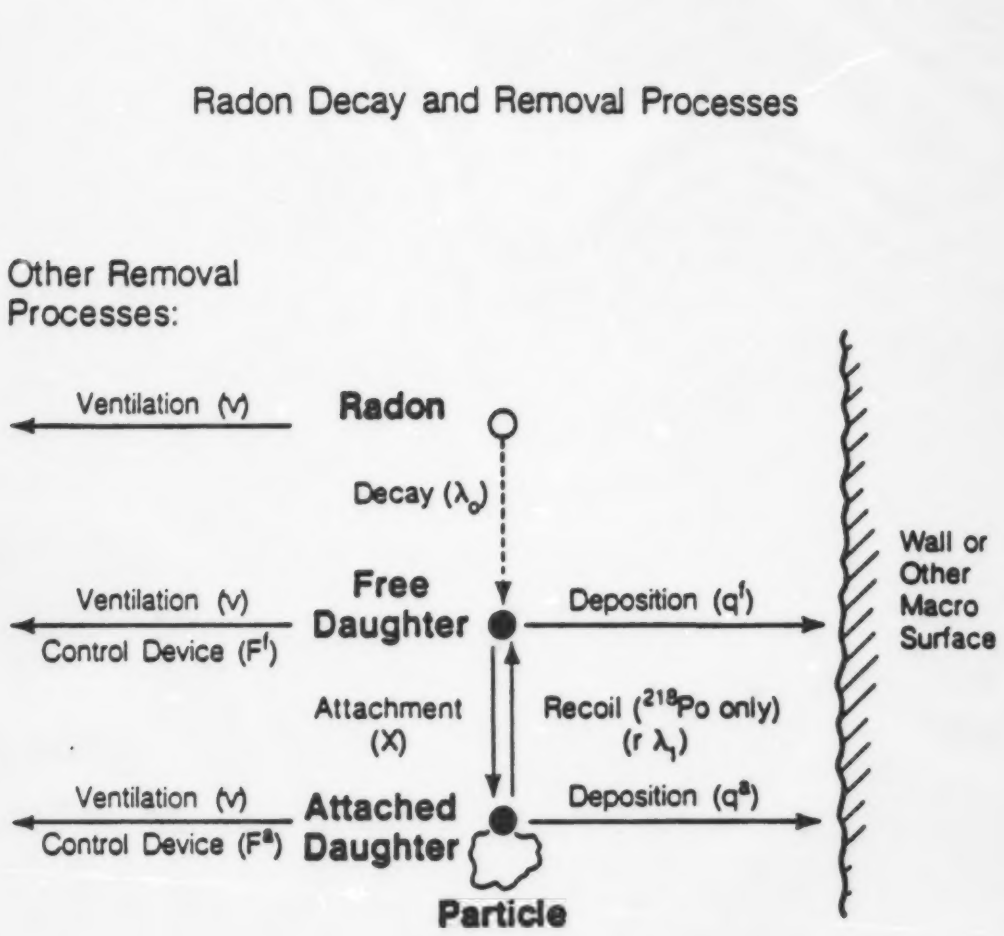

Fig. 12 - Radon decay and removal processes.

APPENDIX A

The following analysis is based on several assumptions:

a) only plane and cylindrical geometries are considered (see Figure A-1);

b) flow of gas takes place perpendicular to the surface of the medium, i.e., one-dimensional problem;

c) the medium through which the gas flows is itself a source of gas;

d) the medium is homogeneous and isotropic.

PLANE GEOMETRY

The geometry of the problem is indicated in Figure A-1. This geometry is more appropriate for dealing with open pit mines and tailings areas. However, the results derived from this simple geometry are sufficiently accurate from a qualitative standpoint to justify this discussion.

The rate of change in the number of radioactive gas atoms in a bulk volume of a porous (ore-bearing) medium where gas production, gas (radioactive) decay, and diffusion and transport of gas take place is given by the continuity condition (1):

$$\frac{\delta C}{\delta t} = -\frac{1}{\eta} \nabla . J_T - \lambda C + S \qquad \text{Eq A-1}$$

where, C is the radioactive gas concentration, number of atoms per unit volume of interstitial space;

J_T is the total current density (flux) of radioactive gas atoms, number of atoms per unit time per unit geometric area 'transported' by molecular diffusion and interstitial fluid (air) flow;

λ is radioactive decay constant of gas;

S is the radioactive gas production rate, number of atoms produced per unit time per unit volume of medium.

η is the porosity of the medium, ratio of void to bulk volume.

In the above Equation J_T can be written as a sum of two terms:

$$J_T = J_D + J_t \qquad \text{Eq A-2}$$

where, J_D and J_t are, respectively, the diffusion and transport current densities. J_D is given by Fick's Law, whereas J_t is determined by Darcy's Law.

The one-dimensional solution of Equation A-1 can now be written as follows:

$$\frac{\delta C}{\delta t} = \frac{D}{\eta}\frac{\delta^2 C}{\delta z^2} - \frac{1}{\eta}\frac{\delta(vC)}{\delta z} - \lambda C + S \qquad \text{Eq A-3}$$

where, v is the transport velocity of the radioactive gas, i.e., velocity of the gas transported by interstitial fluid flow. The z-axis is chosen perpendicular to the mine wall surface, or earth's surface in the case of tailings sites and open pit mines, with the negative direction inward (see Figure A-1). The boundary conditions are:

$$C(z = 0,t) = C(0,t) = 0$$

$$C(z = -\infty,t) = C(-\infty,t) = S/\lambda \qquad \text{Eq A-4}$$

Variables of interest in the context of this paper are the concentration of radioactive gas as a function of wall depth, C(z), and the outward flux of radioactive gas across the wall/air boundary, J_o, the latter being particularly relevant for the calculation of radiation levels in working areas.

Equation A-3 will be solved for a number of cases of interest. Namely:

a) steady-state conditions, i.e., $\delta C/\delta t = 0$, when only diffusion processes are considered and when transport, due to environmental pressure fluctuations, as well as diffusion is taken into consideration;

b) transient behaviour, i.e., $\delta C/\delta t \neq 0$.

STEADY-STATE CONDITIONS

a) No-transport

Equation A-3 for steady-state conditions when transport of gas is ignored reduces to:

$$\frac{\delta^2 C}{\delta z^2} - \frac{\eta\lambda}{D} C + \frac{\eta S}{D} = 0 \qquad \text{Eq A-5}$$

The solution of Equation A-5 subject to the boundary conditions given

by Equation A-4 can be shown to assume the form:

$$C(z) = \frac{S}{\lambda}\left[1 - \exp\left(z(\eta\lambda/D)^{1/2}\right)\right] \qquad \text{Eq A-6}$$

The flux across the wall/air boundary, J_o, can be calculated from Equations A-6 and 1 (see text). The solution is:

$$J_o = -D\nabla C|_{z=0} = S(\eta D/\lambda)^{1/2} \qquad \text{Eq A-7}$$

b) Transport

If transport as well as diffusion are considered to be the steady-state, the equation assumes the form (2):

$$\frac{\delta^2 C}{\delta z^2} - \frac{1}{D}\frac{\delta(vC)}{\delta z} - \frac{\eta\lambda}{D}C + \frac{\eta}{D}S = 0 \qquad \text{Eq A-8}$$

Equation A-8 is difficult to solve analytically even for simple forms of the transport velocity, v. A simplification can be introduced for the particular laboratory controlled condition where a constant, depth independent, transport velocity is established when a suitable external pressure differential is applied between the two end phases of the bulk material. In this case, one may write:

$$\frac{\delta(vC)}{\delta z} \simeq v\frac{\delta C}{\delta z} \qquad \text{Eq A-9}$$

Substituting Equation A-9 into Equation A-8 and solving subject to the boundary conditions indicated by Equations A-4, it can be shown that:

$$C(z) = \frac{S}{\lambda}\left[1 - \exp\left(\frac{v}{2D} + \sqrt{\left(\frac{v}{2D}\right)^2 + \frac{\eta\lambda}{D}}\right) z\right] \qquad \text{Eq A-10}$$

It is easy to see that Equation A-10 reduces to Equation A-6 for $v = 0$.

The flux across the wall/air boundary, taking into account transport and diffusion of gas, J_1, can be calculated from Equations 1 (see text) and A-10. The result is:

$$J_1 = \frac{DS}{\lambda}\left(\frac{v}{2D} + \sqrt{\left(\frac{v}{2D}\right)^2 + \frac{\eta\lambda}{D}}\right) \qquad \text{Eq A-11}$$

Although Equations A-10 and A-11 rarely apply to actual field conditions, they are nevertheless of some value for calculating approximate estimates of C and J. In this heuristic approach, $v = v(t,z)$, is first

calculated and then substituted independently into Equations A-10 and A-11. A more realistic approach than that for the transport velocity assumes different forms according to the kind of barometric pressure variation to which the location of interest is subjected. There are a number of cases of interest, namely: periodic (e.g., sinusoidal) pressure variation and linear pressure variation. In both cases the pressure variation is assumed small compared with its mean value P_o.

Clements and Wilkening (2) considered the case of a transport velocity due to a linear change in the atmospheric pressure P(t) of the form:

$$P(t) = P_o + p(t)$$

where $p(t) = at$ and $p(t) \ll P_o$, and a and t represent, respectively, a constant and time. p(t) is the pressure variation.

Clements and Wilkening give the following expression for the transport velocity:

$$v(z,t) = v(0,t)\ [0.5\ \alpha\pi^{1/2}\ zt^{-1/2}\ \mathrm{erfc}(-0.5\ \alpha z t^{-1/2}) + \exp(-0.25\ \alpha^2 z^2 t^{-1})] \qquad \text{Eq A-12}$$

where, erfc is the complementary error function, and v(0,t) and α are given respectively, by:

$$v(0,t) = v_o = -2a(\eta k/\pi\delta\ P_o)^{1/2}\ t^{1/2}, \text{ and} \qquad \text{Eq A-13}$$

$$\alpha = (\eta\delta/kP_o)^{1/2}$$

For a sinusoidal pressure variation it can be shown that the transport velocity is given by (3):

$$v(z,t) = (ka_oA/\delta)\ \exp(-Az/\sqrt{2})\ \cos(t - Az/\sqrt{2} - \pi/4) \qquad \text{Eq A-14}$$

where, $A = (\eta\ \delta/kP_o)^{1/2}$, and a_o is a numerical constant.

Transport phenomena for radon gas, assuming a sinusoidal pressure variation, have been dealt with by Jeter et al. (4).

From the radiation standpoint, barometric pressure variations of the linear and sinusoidal type are most important in underground mines and uranium tailings piles. Controlled barometric pressure changes can be brought about

in order to affect, in principle, the migration of radon and thoron gases in mine walls. The extent of the pressure-effect on J_1 depends on a number of factors such as the rock porosity and permeability. Variations in the barometric pressure due to large-scale atmospheric pressure changes associated with the passage of frontal systems is also important in tailings sites where uncontrolled environmental conditions prevail. Sudden, abrupt, barometric pressure variations caused by the ventilation system, or as in the case for tailings sites, wind gusts, give rise to slight fluctuations of pressure near the wall or soil surface. Pressure fluctuations of this kind are often described by two terms: an exponentially depth-dependent term, and a sinusoidal time-dependent term.

CALCULATION OF THE RADON TO THORON FLUX RATIO AT THE WALL/AIR INTERFACE

The source of radon or thoron referred to above, i.e., S, can be written as:

$$S = K_I \lambda_I C_I \qquad \text{Eq A-15}$$

where, λ_I and C_I are, respectively, the decay constant and concentration (in atoms per unit volume) of the parent product, i.e., Ra-224 for Rn-220 (thoron) and Ra-226 for Rn-222 (radon). K_I is a dimensionless quantity called the emanation coefficient for Rn-220 or Rn-222. K_I indicates the fraction of the radioactive gas formed by decay of its parent that actually 'escapes' the crystal structures. The rate of emanation of Rn-222 or Rn-220 from a mineral is a function of the structure and uniformity of the mineral crystal lattices, the distribution of the parents Ra-226 or Ra-224, and the degree of branching and the size of capillary networks. The extent of de-emanation, i.e., K_I, is usually less than 100% as a fraction of the radioactive gas formed is trapped irreversibly in crystal structures.

For ores containing U-238 and Th-232, the radon gas to thoron gas flux ratio emanating from the mine walls into the mine atmosphere is given by:

$$\frac{J_{Rn}}{J_{Tn}} = \frac{S_{Rn}}{S_{Tn}} \left(\frac{\lambda_{Tn}}{\lambda_{Rn}}\right)^{1/2} F \qquad \text{Eq A-16}$$

where, the subindices Rn and Tn stand, respectively, for radon gas and thoron gas. (For simplicity it has been assumed that the diffusion coefficients of radon and thoron gas in the medium are the same.) F is a flux ratio function whose value depends on the particular model used. It can be shown that if $D_{Rn} \simeq D_{Tn}$, $\delta_1/\delta_2 \simeq 1$, and $K_{Rn} \simeq K_{Tn}$, the flux ratio becomes:

$$\frac{J_{Rn}}{J_{Tn}} \simeq (3 \times 78)\ F \qquad \text{Eq A-17}$$

where, the numerical value 78 represents the ratio $(\lambda_{Tn}/\lambda_{Rn})^{1/2}$.

Equation A-17 shows that for the plane semi-infinite model the flux of radon gas across the wall/air interface is about 240 times greater than that corresponding to thoron gas. Both fluxes are affected by barometric pressure variations, the effect becoming more pronounced for high porosity media.

REFERENCES

1. Welty, J.R., Wicks, C.E. and Wilson, R.E., "Fundamentals of momentum, heat and mass transfer"; John Wiley and Sons, New York; 1969.

2. Clements, W.E. and Wilkening, M.H., "Atmospheric pressure effects on Rn-222 transport across the earth-air interface"; J. Geophys. Research, vol. 79, No. 33, pp. 5025-5029; 1974.

3. Fukuda, W.. "Air and vapor movement in soil due to wind gustiness"; Soil Science, vol. 79, pp. 249-258; 1955.

4. Jeter, H.W., Martin, J.D. and Schutz, D.F., "The migration of gaseous radionuclides through soil overlying a uranium ore deposit: a modelling study"; Report GJBX-67(77), IWL-4611-410, Teledyne Isotopes; 1977.

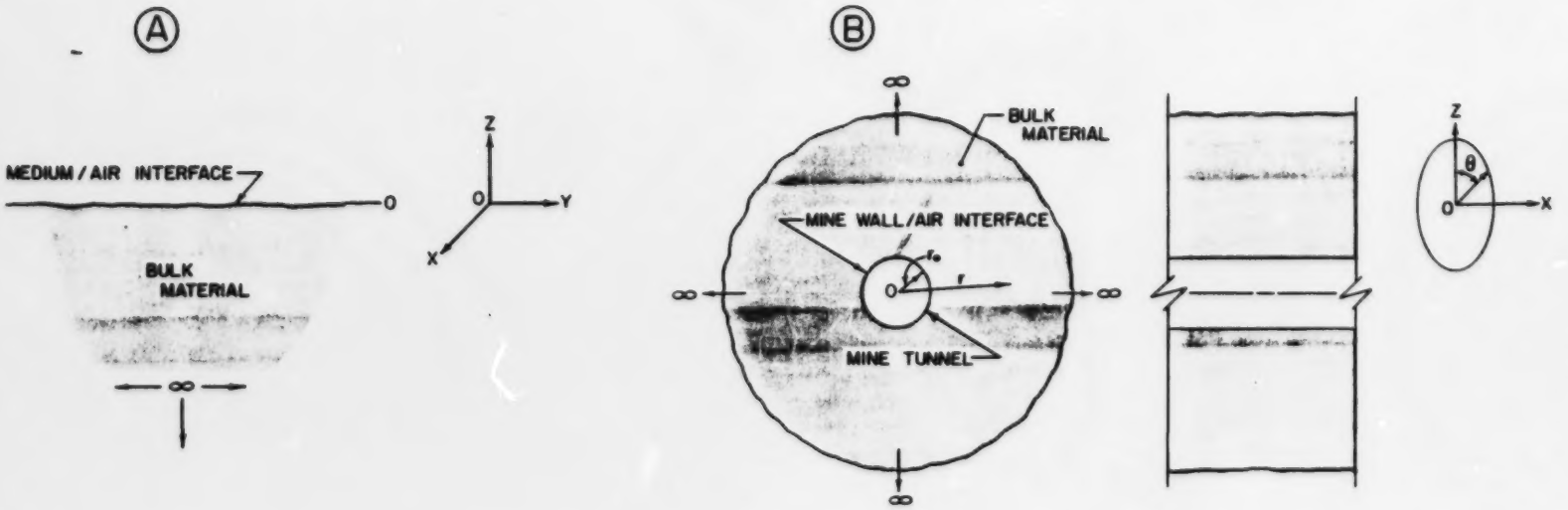

Fig. A-1 - Plane geometry model (drawing A), and cylindrical geometry model (drawing B). Also shown are the coordinate systems used.

APPENDIX B

The expressions for the growth of ^{222}Rn, ^{220}Rn, and their respective progenies, for several (radiation) mine models have been given in detail by Bigu elsewhere (34). Because of the length and complexity of these expressions they are not reproduced here and the interested reader should consult the pertinent references.

The case of a discrete source of ^{222}Rn entering a mine opening is, however, relatively straightforward and will be discussed here. The Mine-Tunnel Model with Ventilation (MTMV) will be used for this purpose (33,34). Assuming that water entering the mine is the source of ^{222}Rn, and calling:

V the volume of a given section of the mine of length L, and circular cross-sectional area, A, of radius R;

Q the ventilation rate, i.e., air flow rate;

$(N_n)_o$ the number of atoms of ^{222}Rn entering the mine at the entry site;

λ_n, λ_A, λ_B and λ_C the radioactive decay constants corresponding to ^{222}Rn, ^{218}Po (RaA), ^{214}Pb (RaB), and ^{214}Po (RaC'), respectively; and assuming the initial conditions at t=0, such that $(N_A)_o = (N_B)_o = (N_C)_o = 0$ (where the indices A, B and C refer, respectively to ^{218}Po, ^{214}Pb, and ^{214}Po, and o refers to t=0), it is easy to show (34) that:

$$N_n(t) = (N_n)_o \, e^{-\Lambda_n t} \quad , \qquad \text{Eq B-1}$$

$$N_A(t) = \frac{(N_n)_o \, \lambda_n}{\Lambda_A - \Lambda_n} \left(e^{-\Lambda_n t} - e^{-\Lambda_A t}\right) \quad , \qquad \text{Eq B-2}$$

$$N_B(t) = \frac{(N_n)_o \, \lambda_A \lambda_n}{\Lambda_A - \Lambda_n} \left[\frac{1}{\Lambda_B - \Lambda_n}\left(e^{-\Lambda_n t} - e^{-\Lambda_B t}\right) + \frac{1}{\Lambda_B - \Lambda_A}\left(e^{-\Lambda_B t} - e^{-\Lambda_A t}\right)\right] \qquad \text{Eq B-3}$$

$$N_C(t) = (N_n)_o \, \lambda_n \lambda_A \lambda_B \left[\frac{e^{-\Lambda_n t} - e^{-\Lambda_C t}}{(\Lambda_A - \Lambda_n)(\Lambda_B - \Lambda_n)(\Lambda_C - \Lambda_n)} + \frac{e^{-\Lambda_A t} - e^{-\Lambda_C t}}{(\Lambda_n - \Lambda_A)(\Lambda_B - \Lambda_A)(\Lambda_C - \Lambda_A)}\right.$$

$$\left. + \frac{e^{-\Lambda_B t} - e^{-\Lambda_C t}}{(\Lambda_B - \Lambda_n)(\Lambda_B - \Lambda_A)(\Lambda_C - \Lambda_B)}\right] \qquad \text{Eq B-4}$$

where, in the above expressions:

$$\Lambda_n = \lambda_n + Q/V$$

$$\Lambda_A = \lambda_A + Q/V \quad , \text{ etc.}$$

It should be noted that specific reference to ^{222}Rn and its progeny (and not to ^{220}Rn and its progeny) has been made in the above expressions. Furthermore, the initial condition $(N_A)_o = (N_B)_o = (N_C)_o = 0$ has been assumed. The reasons for this are as follows:

1. Water entering the mine is the radiation source;
2. ^{222}Rn and ^{220}Rn dissolved by water are carried to the mine site, but because ^{220}Rn (and also ^{222}Rn) originates at locations situated at distances much greater than the diffusion length of ^{220}Rn in the medium, the contribution of ^{220}Rn and hence, its progeny, can be considered negligible;
3. Being metal elements (i.e., radioisotopes of Po, Bi and Pb), the ^{222}Rn and ^{220}Rn progenies formed in water by the radioactive decay of their respective radioactive gas parents will remain in 'solution', and hence, not become airborne.

Finally, a further assumption has been made, namely, all ^{222}Rn dissolved in water is released instantaneously at the mine's entry port. This is, of course, not true, but this oversimplification does not change the qualitative aspect of the results significantly.

The case of multiple discrete ^{222}Rn sources from multiple water entry ports in the mine is more elaborate than the simple case discussed above, but is conceptually straightforward. The final result depends on the number of discrete sources, their strength, and their geometrical distribution within the mine.